OPPORTUNITIES AND LIMITATIONS OF AEROSOL SENSORS TO URBAN AIR QUALITY MONITORING

JOEL KUULA

Opportunities and limitations of aerosol sensors to urban air quality monitoring

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Abstract

Atmospheric particles are one of the leading mortality risk factors in the Global Burden of Disease study (GBD). The association between particulate mass of particles smaller than 2.5 µm in diameter (PM2.5) and cardiovascular and pulmonary diseases has been characterized by multiple epidemiological studies, and varying estimates suggest that several million premature death occur globally each year due to PM2.5 exposure. Mitigation of the adverse health effects of particulate matter requires comprehensive understanding of their sources and dynamic processes, such as spatial dispersion.

Recent emergence and development of aerosol sensors, which are typically characterized as small, relatively low cost and easy to use, have enabled new opportunities in air quality monitoring. As a result of their practical convenience, sensors can be deployed to the field in high quantities which, consequently, enables network-type, spatially comprehensive measurements. However, with more simplified and less expensive measurement approach, less accurate and reliable results may be expected. This study aimed to evaluate and characterize the accuracy and usability of aerosol sensor to urban air quality measurements. The investigation focused on two of the most prominent measurement techniques applicable to sensor type monitoring; optical and diffusion chargingbased techniques. Sensors utilizing optical technique were evaluated in laboratory and field studies for their error sources and particle size-selectivity, specifically. Diffusion charging-based sensors, which measure lung deposited surface area of particles, were evaluated in the field for their suitability to measure combustion emitted particles, such as vehicular exhaust and residential wood combustion emissions.

Results of the study indicated that optical aerosol sensors are unlikely to be fit for long-term regulatory monitoring. The main issues preventing this arise from their improper calibration which poses a significant risk of data misinterpretation; none of the laboratory evaluated sensors measured particle sizes which their technical specifications implied. On the other hand, field tests showed that when the measured size fraction was targeted to match the true detection range of the sensor, highly accurate and repeatable results were obtained. This implies that, while the usability of optical sensors is limited in their current form, the concept and vision of a sensor driven air

quality monitoring network remains valid and achievable. In comparison to optical sensors, diffusion charging-based sensors were found to be more mature in terms of their technological development. The evaluated sensors exhibited accurate and stable performance throughout the test campaigns and were shown to be particularly well-suited the measurement of combustion emitted particles. Hence, diffusion charger sensors would be a valuable addition to be used alongside other measurement techniques as urban air quality is heavily affected by nanoparticles.

Tekijä

Joel Kuula

Nimeke

Aerosolisensoreiden mahdollisuudet ja rajoitteet kaupunki-ilmanlaadun seurannassa

Tiivistelmä

Ilmakehän pienhiukkaset ovat yksi keskeisimmistä kuolleisuuden riskitekijöistä kansainvälisessä taudin rasittavuuden analyysissä. Useat epidemiologiset tutkimukset ovat osoittaneet pienhiukkasten ja sydän- ja verisuoni- sekä hengitystiesairauksien yhteyden, ja eri arvioiden mukaan useita miljoonia ennenaikaisia kuolemia tapahtuu joka vuosi pienhiukkasaltistumisen seurauksena. Jotta pienhiukkasten negatiivisiin terveysvaikutuksiin voitaisiin vaikuttaa, tulee niiden lähteet ja dynaamiset prosessit, kuten alueellinen leviäminen, tuntea hyvin.

Viimeaikainen aerosolisensoreiden esilletulo ja kehittyminen ovat avanneet uusia mahdollisuuksia ilmanlaadun seurantaan. Sensorit, jotka ovat tyypillisesti pienikokoisia, suhteellisen edullisia ja helppokäyttöisiä, mahdollistavat alueellisesti kattavien sensoriverkkomittausten toteuttamisen ja siten pienhiukkasten tarkemman tutkimisen. Sensoreiden edullisempi ja siten yksinkertaisempi mittaustekniikka saattaa toisaalta johtaa suurempaan mittausepätarkkuuteen ja huonompaan laatuun. Tämän työn tavoitteena oli arvioida ja luonnehtia aerosolisensoreiden tarkkuutta ja soveltuvuutta kaupunkialueiden ilmanlaadun seurantaan. Tutkimus keskittyi kahteen mittaustekniikkaan, jotka ovat parhaiten sovellettavissa sensorityyppisiin mittauksiin; optiseen ja diffuusiovarautumiseen perustuvaan tekniikkaan. Optisia sensoreita testattiin sekä ulkoilmassa että laboratoriossa, missä niiden hiukkaskokovalikoivuutta arvioitiin tutkimalla sensorin vastetta keinotekoisesti tuotetuilla erikokoisilla referenssihiukkasilla. Diffuusiovarautumiseen perustuvia sensoreita, jotka mittaavat niin kutsuttua keuhkodeposoituvaa pinta-ala, testattiin ulkoilmassa, missä niiden suorituskykyä arvioitiin erityisesti erittäin pienten nanohiukkasten, kuten liikenteen pakokaasun sekä puunpolton päästöjen, näkökulmasta.

Tutkimustulosten perusteella optiset aerosolisensorit eivät toistaiseksi ole soveltuvia pitkäaikaiseen viranomaisvalvonnassa tehtävään ilmanlaadun seurantaan. Tämä johtuu niiden virheellisestä kalibroinnista, jonka seurauksena sensorit eivät mittaa hiukkaskokoluokkia, joita niiden tekniset tuoteselosteet antavat olettaa. Riski mittausdatan väärin tulkinnalle on täten ilmeinen. Toisaalta, kun mitattu hiukkasten kokojakauma rajattiin vastaamaan sensorin ominaista vastealuetta, sensorin mittaustarkkuus oli hyvä ja toistettava. Tämän perusteella, vaikkakin virheellinen kalibrointi rajoittaa optisten sensoreiden käytettävyyttä, konsepti ja visio sensoripohjaisesta mittausverkosta on mahdollinen ja saavutettavissa. Diffuusiovarautumiseen

perustuvat sensorit osoittivat olevan teknisesti kehittyneempiä kuin optiset sensorit. Testatut sensorit olivat tarkkoja ja stabiileja kaikissa kenttämittauskampanjoissa, ja ne olivat erityisen hyvin soveltuvia liikenteen pakokaasujen sekä puunpolton päästöjen mittaamiseen. Tämän vuoksi diffuusiovaraukseen perustuvat sensorit olisivat arvokas lisä muiden mittaustekniikoiden rinnalle, varsinkin kun nanohiukkasten osuus kaupunki-ilmassa on merkittävä.

Acknowledgements

Contents

List of Abbreviations and Symbols

APS   Aerodynamic particles sizer

BC   Black carbon

BrC   Brown carbon

CCN   Cloud condensation nuclei

CFD   Computational fluid dynamics

CMD   Count median diameter

CPC   Condensation particle counter

CV   Coefficient of variation

DH   Detached housing area station

DMA   Differential mobility analyser

DMPS   Differential mobility particle sizer

DOS   Dioctyl sebacate

FEM   Federal equivalent method

FMI   Finnish Meteorological Institute

GDP   Gross domestic products

GSD   Geometric standard deviation

HSY   Helsinki Region Environmental Services Authority

ICRP   International Commission on Radiological Protection

IPCC   Intergovernmental Panel on Climate Change

LDSA   Lung deposited surface area

LPO   Lo pulse occupancy

MMD   Mass median diameter

NOx   Nitrogen oxide

NPF   New particle formation

NRMSE	Normalized root mean square error
OEM	Original equipment manufacturer
OPC	Optical particle counter
PA	Palmitic acid
PAS	Prototype aerosol sensor
PM	Particulate matter
PM10	Particulate mass < 10 μm
PM2.5	Particulate mass < 2.5 μm
PM2.5-10	Particulate mass 2.5 – 10 μm
RMSE	Root mean square error
RSD	Relative standard deviation
SC	Street canyon station
SCAQMD	South Coast Air Quality Management District
SMEAR	Station for Measuring Ecosystem—Atmosphere
SMPS	Scanning mobility particle sizer
SOA	Secondary organic aerosol
TEOM	Tapered element oscillating microbalance
UB	Urban background station
VOAG	Vibrating orifice aerosol generator

C	Volumetric concentration
$\bar{c}_i$	Mean thermal speed of ions
d_d	Droplet diameter
d_p	Particle diameter
e	Charge of an electron
E_{ch}	Particle charging efficiency
ε	Relative permittivity of a particle
f	Frequency
i	ith measurement point
I	Volumetric fraction of impurity
k	Boltzmann's constant

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K_E	Electrostatic constant of proportionality
N_i	Concentration of ions
Q	Liquid flow rate
R_{APS}	Response signal of the APS (total mass)
R_s	Response signal of the sensor
S_{LDSA}	Lung deposited surface area of a sphere-shaped particle
T	Temperature
t	Time
Z_i	Mobility of ions

List of Publications

This doctoral dissertation consists of a summary and of the following publications which are referred to in the text by their numerals

1. Kuula, Joel; Mäkelä, Timo; Hillamo, Risto; Timonen, Hilkka. 2017. Response characterization of an in inexpensive aerosol sensor. Sensors, volume 17, issue 12, doi: 10.3390/s17122915.

2. Kuula, Joel; Mäkelä, Timo; Aurela, Minna; Teinilä, Kimmo; Varjonen, Samu; González, Óscar; Timonen, Hilkka. Evaluation of particle size-selectivity of optical low-cost particulate matter sensors. Submitted to the journal of Atmospheric Measurement Techniques in the year 2019, doi: 10.5194/amt-2019-422

3. Kuula, Joel; Kuuluvainen, Heino; Rönkkö, Topi; Niemi, Jarkko V.; Saukko, Erkka; Portin, Harri; Aurela, Minna; Saarikoski, Sanna; Rostedt, Antti; Timonen, Hilkka. 2019. Applicability of optical and diffusion charging-based particulate matter sensors to urban air quality measurements. Aerosol and Air Quality Research, volume 19, issue 5, pages 1024-1039, doi: 10.4209/aaqr.2018.04.0143.

4. Kuula, Joel; Kuuluvainen, Heino; Niemi, Jarkko V.; Saukko, Erkka; Portin, Harri; Kousa, Anu; Aurela, Minna; Rönkkö, Topi; Timonen, Hilkka. 2019. Long-term sensor measurements of lung deposited surface area of particulate matter emitted from local vehicular and wood combustion sources. Aerosol Science and Technology, volume 53, issue 11, doi: 10.1080/02786826.2019.1668909

Author's Contribution

Publication 1: Response characterization of an inexpensive aerosol sensor

The author co-designed the experiment setup together with Timo Mäkelä and conducted the experiments. The author designed and assembled the sensor system, analysed the experiment results, and wrote the publication with the help and feedback from all co-authors. Rationale for the research was drafter together with Risto Hillamo and Hilkka Timonen.

Publication 2: Laboratory evaluation of particle size-selectivity of optical low-cost particulate matter sensors

The author co-designed the experiment setup together with Timo Mäkelä and conducted the tests. The author designed and assembled the sensor systems and analysed the results. The author wrote the publication with the help and feedback from all co-authors. University of Helsinki and Fab Lab Barcelona provided some of the sensors used in the evaluations.

Publication 3: Applicability of optical and diffusion charging-based particulate matter sensors to urban air quality monitoring

The author designed and assembled the optical sensor system and was responsible for its operation during the measurement campaign. Diffusion charging-based sensors were operated by Heino Kuuluvainen and by the HSY staff. Reference instrumentation was operated and maintained solely by the HSY staff. The author analysed the results and wrote the publication with the help and feedback from all co-authors. A section in the publication regarding state-of-the-art description of diffusion charging-based sensors and Pegasor AQ Urban was first drafted by Heino Kuuluvainen, Topi Rönkkö, Antti Rostedt, and Erkka Saukko.

Publication 4: Long-term sensor measurements of lung deposited surface area of particulate matter emitted from local vehicular and wood combustion sources

The measurements were carried out under the leadership of HSY. The author analysed the results and wrote the publication with the help and feedback from all co-authors. Pegasor AQ Urban device description was written drafted by Heino Kuuluvainen with the support of Erkka Saukko.

1. Introduction

1.1 Background

Recent emergence and development of aerosol sensors has enabled new opportunities in air quality monitoring (Morawska et al. 2018; Kumar et al. 2015; Snyder et al. 2013). Whereas the conventionally used instrumentation has often been perceived as expensive and impractical in several different ways (e.g. Brauer et al. 2019), the sensor based complementary approach is predicated on the relatively low unit cost, compact size, and ease of use of the sensors. These features make it possible to cost-effectively extend the coverage of measurements and thus achieve better understanding of the dynamic variations of particulate matter (PM). Especially in urban areas, PM can be highly heterogeneously distributed due to several different local sources being simultaneously present within a relatively small area (Eeftens et al. 2015). Therefore, a single measurement point would only represent the specific conditions found at that specific location. By extending the spatial coverage of standardized instruments with lower and easier to use sensors, benefits could be achieved, for example, in the development of dispersion models and forecasting of air quality (Popoola et al. 2018; Johansson et al. 2015). However, it is also important to note that with more simplified and thus less expensive measurement methods, less accurate and reliable results may be expected.

The importance of distributed sensing and detailed spatial characterization of ambient PM is highlighted by its propensity to inflict adverse health effects; PM is among the top ranking mortality risk factors in the global burden of disease analysis, and varying estimates suggest that several million premature deaths are attributable to PM exposure globally each year (Burnett et al. 2018; Cohen et al. 2017; Lelieveld et al. 2015). Furthermore, along with adverse health effects and their societal impacts on economy, ambient aerosols have direct and indirect climatic effects which, in comparison to the climatic effects of greenhouse gases, still exhibit large uncertainties (IPCC 2013).

Atmospheric particles are diverse in their physiochemical properties (Hinds 1999). Depending on their formation and aging processes, their size (diameter) may vary from nanometers to hundreds of micrometers implying a difference of ~ 5 in the order of magnitude. Other properties such as particle shape, density, and chemical composition and volatility are also subject to variation. Partly due to these factors, measurement of particles is particularly challenging, and typically several different measurement techniques must be utilized in parallel in

order to obtain comprehensive understanding of the prevailing conditions. Amongst sensors, the most prominent techniques are based on optical detection of light scattering from illuminated particles, and on the unipolar diffusion charging of particles and consequent measurement of electric current. Both techniques have different strengths and weaknesses.

1.2   Objectives

This study investigates the opportunities and limitations of PM sensors to monitoring of urban air quality. In particular, the research is focused on the characterization of error sources of optical sensors regarding their particle size-selectivity (Publications 1, 2, and 3) and, respectively, on the evaluation of the suitability of diffusion charging-based sensors to the measurement of combustion emitted particles (Publications 3 and 4). A demonstration how a group of sensors can be utilized in the assessment of local variability of PM is also made (Publication 4). Together these publications aim to provide insight to whether aerosol sensors, as a novel methodological approach, can be used to support and advance current technical and scientific understanding of atmospheric particles and their measurement.

1.3   Limitations

The main limitation of this study is the lack of comprehensive sensor network experiments. Deployment of a massive-scale sensor network is a substantial effort and requires not only on-going maintenance and upkeep but also sophisticated infrastructure for data transfer and data processing. This, in turn, requires exceptionally comprehensive resources which, in the scope of this thesis, were not available. Thus, although the performance of a single sensor can be measured and quantified accurately in laboratory and field evaluations, it remains unclear to what degree the utilization of a massive-scale sensor network would improve understanding of the features and properties of city-scale air quality dynamics. Related to this, analysis of the unforeseen issues and problems, which the deployment of networks and widespread unfolding of sensor type measurements may entail, are also out of the scope of this study. Future efforts should still, nevertheless, be aimed for the development and deployment of large networks as the obtained data would be unprecedented and of great interest to the atmospheric science community.

A minor limitation of the study is that it does not investigate the effect of relative humidity on sensor accuracy. In spite the fact that current literature suggests relative humidity and consequent hygroscopic growth of particles being a significant source of distortion in the sensor responses, it is a universal problem and not intrinsically related to aerosol sensors only.

1.4 Scientific contribution

Publications 1 and 2 outline the development and utilization of a novel reference aerosol generation method, which enables comprehensive examination of particle size-selectivity of optical sensors. While existing studies have noted and verified that the sensor response is affected by the differing particle sizes, they have not been able to identify and quantify their complete detection range. To the author's knowledge, the evaluation approach, which is described in Publications 1 and 2, and the respective results, which provide clear evidence to the notion that particle size-selectivity has an essential role in the error source analysis of optical sensors, are unprecedented. According to the results, none of the evaluated sensors adhered to the detection ranges their technical specifications sheets implied, although some size bins of some of the individual sensors were relatively close to the stated values. The improper calibration by the manufacturers poses a significant risk of data misinterpretation which, consequently, poses limitations on the sensor usability.

Publication 3 shows how the laboratory results obtained in Publications 1 and 2 manifest themselves in field conditions. Although many optical sensor studies have been conducted in the field, in most cases the evaluation has been done by comparing the sensor output to the standard PM size fractions, such as the PM2.5 or PM10 (mass of particles in < 2.5 and < 10 µm size ranges, respectively), without the detailed knowledge of the sensor detection range. This study showed that if the evaluation is done by comparing the sensor response to a size fraction, which is closer to the characteristic detection range of the sensor (in this case PM2.5-10 instead of PM10) more accurate results can be achieved. In general, the applicability of an optical sensor to urban air quality measurements is dictated by the sensor's valid detection range and how it compares to the size distribution of the measured aerosol. The changing ambient conditions and the limited operational range, which the evaluated sensors appear to exhibit, will inevitably result in trade-offs in the measurement accuracy. This underlines the importance of the sensor user to properly understand and acknowledge the specific characteristics and respective limitations of the used sensor.

Publication 3 also demonstrates an evaluation and inter-comparison of three different diffusion charger sensors and assesses their applicability to the measurement of combustion emitted particles (in this case particles originating from vehicular exhaust emissions). The results are valuable addition to the currently sparse literature and moreover, strengthen the perception that diffusion charging-based measurement technique, in general, is an accurate and reliable method for the measurement of lung deposited surface area. The sensitivity and suitability of LDSA sensors to the measurement of combustion emitted particles indicates that these sensors are a worthwhile addition to urban air quality assessments.

Publication 4 demonstrates how a group of four diffusion charging-based sensors can be utilized in a functional monitoring network. To the author's knowledge, similar experiment has not been conducted previously. In the network, the four sensor systems were placed to three distinctively different meas-

urement sites found in urban areas: street canyon, urban background, and detached housing area. The results showed significant site-specific differences in measured lung deposited surface area concentrations, even in cases where the stations were closely located (two detached housing area stations 670 m apart, urban and urban background stations less than 1 km apart). The stations also exhibited different characteristics regarding diurnal cycles and seasonality. The results underline the utility and need of dense monitoring networks if more accurate air quality assessments are desired.

2. Atmospheric aerosols

2.1 Particulate matter

Term 'aerosol' refers to suspension of solid particles or liquid droplets in a gas medium, such as atmospheric air. Analogous to 'aerosol', another commonly used term is 'particulate matter' or 'PM'. The size of a particle or droplet is determined by its diameter, and more specifically, typically an aerodynamic diameter is used. Aerodynamic diameter refers to the diameter of a particle which has the same settling velocity as a spherical particle with a density of 1000 kg m^{-3}. Aerodynamic diameter standardizes particle shape and density and, therefore, it has found wide application in aerosol technology. In the atmosphere, particle sizes may vary from few nanometres (nm = 10^{-9} m) to hundreds of micrometres (μm = 10^{-6} m) but research is typically focused on particles smaller than 10 μm due to their higher relevance in atmospheric processes and human health effects. Aerosol concentrations are typically measured in number (# cm^{-3}) or mass (μg m^{-3}) basis, but surface area (μm^{2} cm^{-3}) is also used occasionally. Furthermore, particularly in health studies, inhaled doses may be calculated by weighing the particle size distribution of the measured concentration with different respiratory tract deposition fractions.

Urban aerosol is a complex and dynamic mixture of different sized particles emitted and formed from different types of sources. In comparison to gases, such as carbon dioxide, particles exhibit relatively short lifespan (from hours to days as opposed to several or even tens of years) and due to the multitude of different sources, their impact to air quality is highly depend on time and location. Particles originating from direct sources, such as vehicular exhaust emissions, are called primary particles whereas particles formed in the atmosphere via gas-to-particle conversion of biogenic and anthropogenic precursor gases are called secondary particles. Both primary and secondary particles are subject to size-dependent processes of growth (condensation and coagulation), evaporation, and removal, and the particle size distributions, which are observed in the ambient air, are manifestations of these processes acting upon the particles. A typical particle size distribution found in an urban area contains three distinctive modes; nuclei, accumulation, and coarse particle modes (Figure 1, adapted from Hinds, 1999).
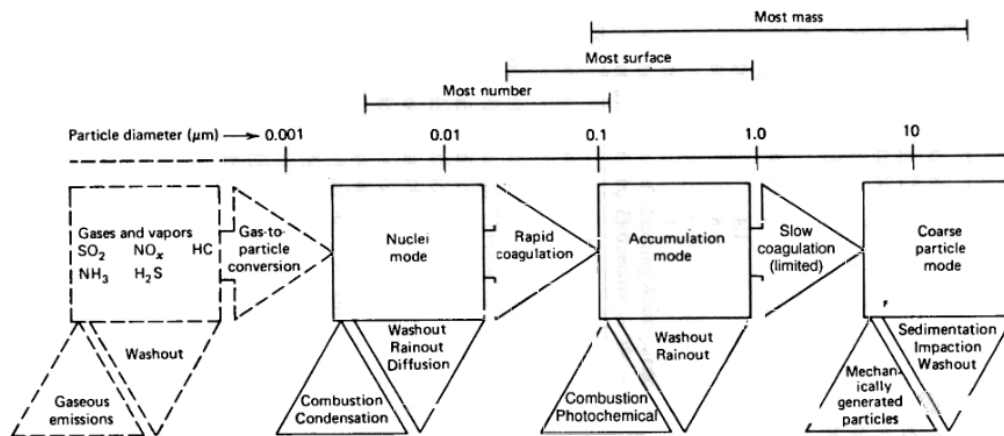
Figure 1. Processes affecting atmospheric particles (adapted from Hinds, 1999).

Nuclei mode (~ 1 – 25 nm) consists of particles resulting from fresh combustion and nucleation (gas-to-particle conversion) (Rönkkö et al. 2017; Brines et al. 2015; Kulmala et al. 2013). These particles are primarily in the sub 10 nm size range, but due to the very small size, they undergo rapid coagulation and form larger particles in a matter of hours. Their lifetime in the atmosphere is short. Nuclei mode is particularly distinct, for example, in road and highway sites where vehicular traffic is in immediate proximity to the measurement station. Accumulation mode (~ 50 nm – 1000 nm) particles are formed in the coagulation of nuclei mode particles, but they are also emitted directly to atmosphere by e.g. residential wood combustion (Tissari et al. 2008). Removal mechanisms for the accumulation mode particles are weak, as the name suggests; they are too large for rapid coagulation and diffusional deposition and too small for effective gravitational washout. Coarse mode particles (> 2.5 µm) consists of mechanically generated particles (e.g. abrasion of tire, dirt, and pavement), windblown dust, and sea salt. Their lifetime in the atmosphere is short due to their heavy weight. Coarse mode particles compose of crustal materials and their oxides whereas fine particles (nuclei and accumulation mode) are typically composed of e.g. hydrocarbons, soot, and sulphates. Concentration wise, an important distinction is made between nuclei mode and accumulation and coarse mode particles. Particle number concentrations are typically dominated by nuclei mode particles, however, their contribution to the total particulate mass is only few percentiles (Figure 2, adapted from Hinds, 1999).

Figure 2. Generalized representation of the magnitude of different particle modes in terms of their particle number concentration (a) and volume concentration (b) (adapted from Hinds, 1999).
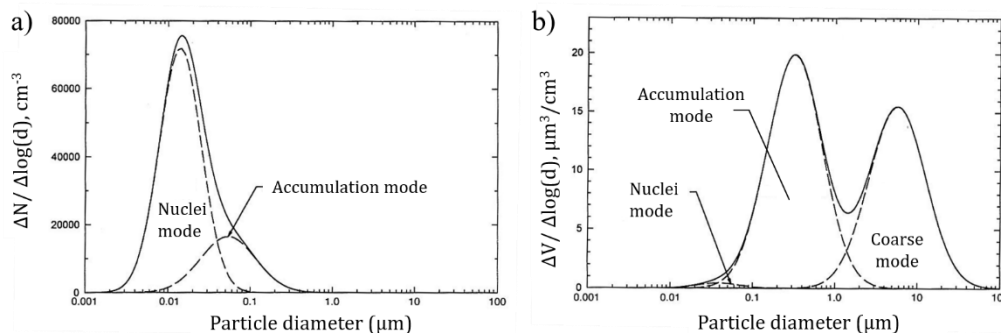
2.2   Health and climatic effects of aerosols

Particulate matter (PM) has been known for causing adverse health effects to humans for several decades (Dockery et al. 1993). The association between the particulate mass of particles smaller than 2.5 μm in diameter (PM2.5) and cardiovascular and pulmonary diseases has been characterized by multiple epidemiological studies (e.g. Hoek et al. 2013; Silva et al. 2013; Brook et al. 2010), and it has been estimated that several million premature deaths occur globally each year due to PM exposure (Burnett et al. 2018; Cohen et al. 2017; Lelieveld et al. 2015). In the EU alone, an estimated 391,000 premature deaths occurred in 2015 (European Environment Agency 2018). In addition to being a tragedy of the individual, premature deaths and illnesses entail far reaching impacts on national economies as well due to their associated healthcare expenditures and losses in gross domestic products (GDP) (Wu et al. 2017; Xie et al. 2016). Despite the vast amount of data and dedicated effort, the underlying processes governing the degrading health effects of PM remain still, to some degree, poorly understood (Kelly and Fussell 2012).

The evidence for the adverse health effects of PM2.5 is clear, however, it is not evident whether it is the most accurate health impact predictor as several other factors, such as particle number and surface area concentration and particle size, chemical composition, water-solubility, and volatility, may also have a significant contribution to the harmfulness of an aerosol. Partly because of this, it has been considered that a parameter known as lung deposited surface area (LDSA) could potentially be a more accurate health predictor alternative. LDSA describes the surface area of particles depositing to the deepest (i.e. alveolar) region lungs, and consequently, it takes into account not only the surface area but also the potential of different toxic and non-toxic chemicals to be translocated into the blood circulation system. Measurement wise, LDSA is an intermediary parameter which cannot be simply inferred from either particle number or mass concentration (Baldauf et al. 2016). LDSA concentrations are mainly driven by particles smaller than 400 nm due to their favourable lung deposition fraction characteristics (Asbach et al. 2009), and in practice, much of the LDSA in urban areas originates from vehicular exhaust emissions and

residential wood combustion. Several studies have shown the propensity of LDSA to inflict oxidative stress and proinflammatory responses (Oberdörster et al. 2005; Brown et al. 2001), but unlike the mass based metrics (PM2.5 and PM10), LDSA remains unregulated and hence largely unmonitored.

Aerosols have a significant effect on climate, as seen in Figure 3 (IPCC 2013). Whereas the greenhouse gases absorb sunlight and warm climate, aerosols scatter sunlight back to the space and thus cool the atmosphere (Charlson et al. 1992). An exception to this are particles of black and brown carbon (BC and BrC, respectively), mineral dust, and some metals which absorb radiation and reduce the planetary albedo effect (Moosmüller et al. 2009). Besides these direct radiative forcing effects, aerosols have indirect climatic effects of forming and adjusting clouds (Lohmann and Feichter 2005). Particles can act as a cloud condensation nuclei (CCN) which accelerate cloud formation. They may also modify the size of cloud particles and thus affect the optical properties of clouds. Overall, aerosols are estimated to have a net cooling effect on the atmosphere, but large uncertainties still exist.
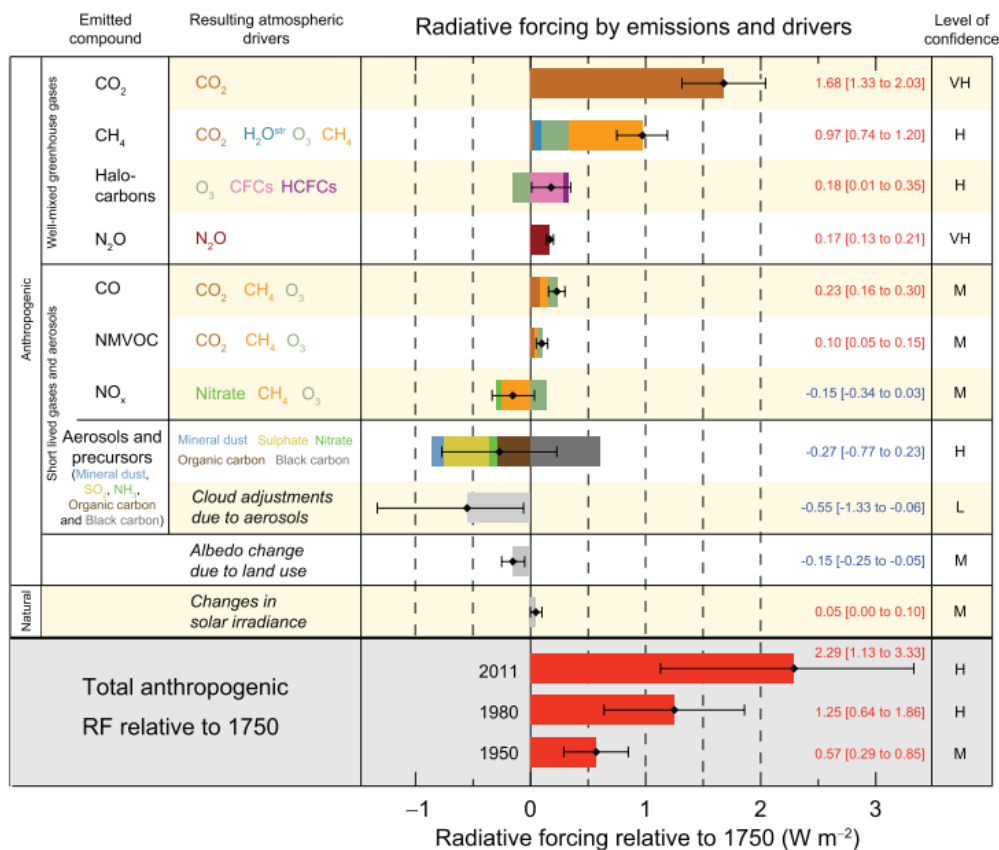

Figure 3. Estimated radiative forcing's of different atmospheric drivers (adapted from IPCC 2013).

2.3 Optical measurement technique

Amongst sensors, the most commonly utilized measurement technique is based on the optical detection of light scattered from an illuminated particle. This detection method is relatively versatile regarding its valid particle detection

range, and a sensor of this type can be designed to be fairly simple with respect to its basic structure. A typical sensing arrangement consists of a light source-photodetector pair, as shown Figure 4 (Grimm and Eatough 2009), in which the light source is positioned in an angle (normally 90-120°) with respect to the photodetector. Light scattering from the particles passing through the beam is sensed and transformed into an electronic voltage signal by the photodetector. Depending on how the sensor has been designed, the electronic signal can be interpreted for its pulse frequency (number of particles) and respective pulse height (size of particles), or alternatively, for its total scattered light intensity resulting from an assembly of particles. A sensor using a single particle signal interpretation (pulse frequency and height) is called optical particle counter (OPC) whereas a sensor measuring total scattered light intensity is called photometer.
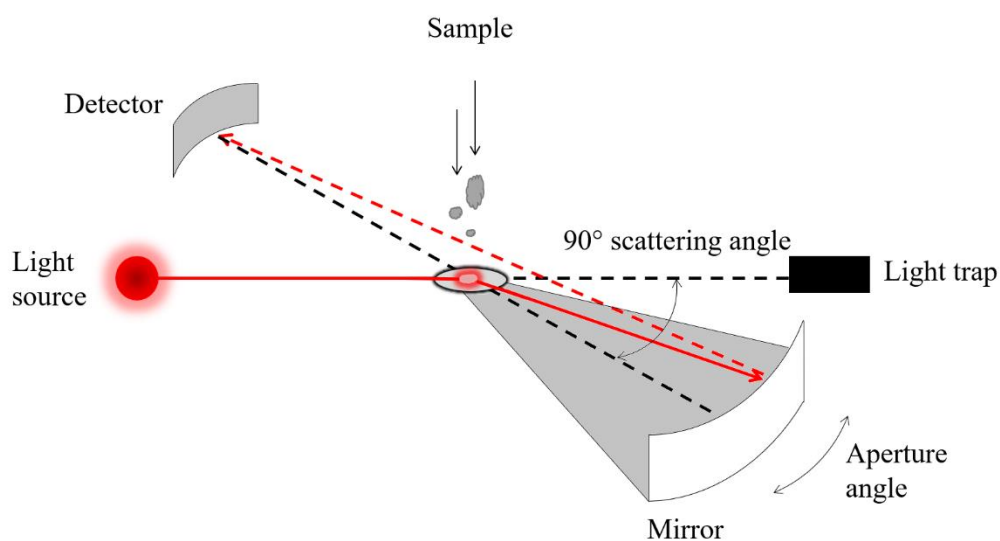
Figure 4. Working principle of a generalized optical particle counter (Grimm and Eaton 2009).

The main weakness of the optical measurement technique is related to the exponentially diminishing light scattering intensity of small particles. By default, optical methods cannot be used to measure ultrafine (< 100 nm) particles as the light scattering intensity of these particles becomes undistinguishable from background noise. Many of the commercial instruments, including sensors, are limited to detection of particles larger than ~ 300 nm (Kulkarni et al. 2011), and in practice, this implies that particles emitted from combustion sources are, for the most part, out of their detection range. This is a major deficit considering that, especially in urban areas, air quality is heavily affected by particles emitted from local vehicular exhaust emissions and residential wood combustion (Glasius et al. 2018; Helin et al. 2018; Rönkkö et al. 2017). To address combustion emitted particles, a diffusion charging-based measurement technique may be used.

2.4   Diffusion charging-based measurement technique

Diffusion charging measurement technique relies on the electrical charging of particles, and an exemplary working schematic of this is shown in Figure 5

(Amanatidis et al. 2016). A high voltage corona is used to form ions, which are driven onto the surfaces of particles by diffusional and field charging forces. Diffusional charging, which is more effective for smaller particles (< 0.1 µm), results from the thermal diffusion-induced Brownian motion of ions and particles and from their subsequent random collisions. Field charging, which is more relevant for larger particles (> 1 µm), occurs when ions exhibit movement in an electric field and collide with particles. The electric field — either an external field or a field formed by the ions — is distorted by the neutrally charged particle, which causes the movement trajectories of ions to coincide with the particle. The level of distortion is dependent on the permittivity of the particle material and, more importantly, on the level of the charge of the particle; the greater the acquired charge, the stronger the repulsive forces between the particle and ions become (i.e. charging saturates). After the particles have been fully charged, excess ions are removed from the sample stream using an ion trap, and an escaping current (i.e. the number of ions attached to the surfaces of particles), is measured at the outlet of the sensor. Alternatively to the measurement of escaping current, particles, and the charge carried by them, may be collected to a Faraday cup filter and measured using an electrometer.
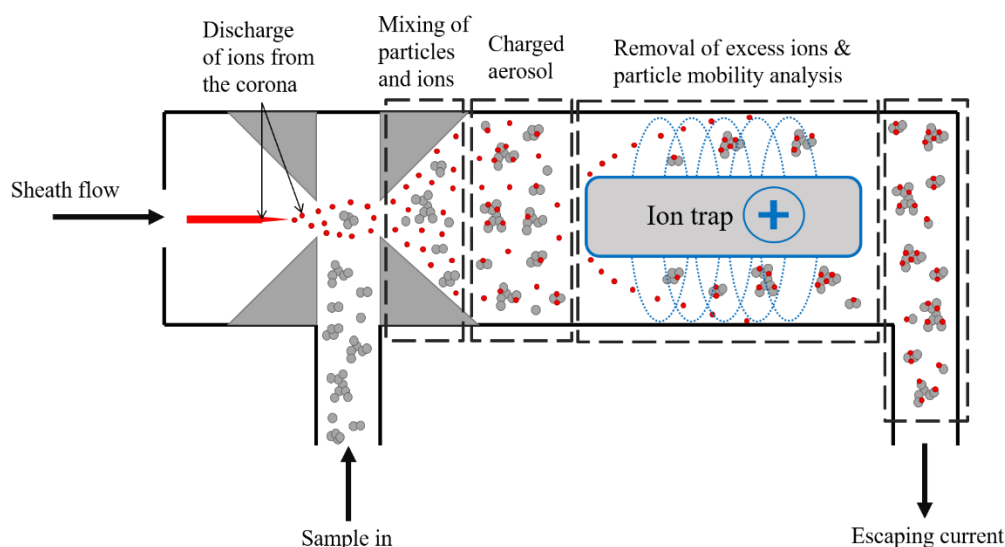
Figure 5. Working principle of a diffusion charging-based Pegasor PPS-M sensor (Amanatidis et al. 2016). Red dots represent ions and grey agglomerates represent particles.

A device utilizing diffusion charging technique is best suited for the measurement of lung deposited surface area (alveolar region) of particles, but its output signal can be converted to correspond number or mass concentration as well. However, this requires that the measured size distribution contains only particles smaller than 400 nm in size, which under typical outdoor conditions, is rarely the case. Number or mass concentration measurements are feasible only in specific cases, for example in automotive exhaust emission measurements (Amanatidis et al. 2016, 2017; Maricq 2013; Ntziachristos et al. 2013).

The suitability of a diffusion charger-based sensor to the measurement of LDSA results from the similar particle size-dependency of particle charging ef-

ficiency and alveolar surface area, as shown in Figure 6. In this figure, an idealized particle charging efficiency (α $d_p^{1.13}$) is plotted in black line, LDSA per particle in red (particle surface area α d_p^2), and particle alveolar deposition fraction in blue line. In respective order, the plotted curves are expressed mathematically as (Equation 3 adapted from Hinds, 1999):

$$E_{ch} = 0.029 * d_p^{1.13} \tag{1}$$

$$S_{LDSA} = DF_{AL} * \pi d_p^2 \tag{2}$$

$$DF_{AL} = \frac{0.0155}{d_p}(e^{\left(-0.416(\ln(d_p)+2.84)^2\right)} + 19.11e^{(-4.82(\ln(d_p)-1.362)^2)}) \tag{3}$$

Where E_{ch} is the particle charging efficiency, d_p is the particle diameter, S_{LDSA} is the lung deposited surface area of a single sphere-shaped particle, and DF_{AL} is the particle alveolar deposition fraction. The constant of 0.029, shown in Equation 1, adjusts the offset of the particle charging efficiency, and it is used here to fit the particle charging efficiency and particle lung deposited surface area curves to similar scale.

Figure 6 shows that, despite the apparent discrepancy in particle size-dependencies of particle charging efficiency and particle surface area ($d_p^{1.13}$ and d_p^2, respectively), the alveolar deposition efficiency modulates the particle surface area in a way that, if an error margin of $\pm$ 30 % is tolerated, a nearly linear relationship is formed between the two in the size range of ~ 20 – 400 nm. Although the earliest versions of diffusion chargers were developed in 1970-1980s (Lehtimäki 1983; Liu and Lee 1976) this connection between the LDSA and particle charging efficiency was only discovered in 2007 by Wilson et al. The particle charging efficiency of $d_p^{1.13}$, used in Equation 1, is empirically derived (Jung and Kittelson 2005) and other values have also been presented; 1.034 (Bau et al. 2012), 1.099 (Bau et al. 2015), 1.11 (Fierz et al. 2007), 1.125 (Fierz et al. 2011), and 1.16 (Wilson et al. 2007). The value represents the combined effect of the diffusional and field charging processes. Mathematical expressions of these processes (Equations 4 and 5, respectively) have been shown by Hinds (1999):

$$n(t) = \frac{d_p kT}{2K_E e^2} \ln\left(1 + \frac{\pi K_E d_p \bar{c}_i e^2 N_i t}{2kT}\right) \tag{4}$$

$$n(t) = \left(\frac{3\varepsilon}{\varepsilon+3}\right)\left(\frac{E d_p^2}{4K_E e}\right)\left(\frac{\pi K_E e Z_i N_i t}{1+\pi K_E e Z_i N_i t}\right) \tag{5}$$

Where n is the number of charges acquired by a spherical particle d_p after time duration t, k is the Boltzmann's constant, T is the temperature, K_E is the electrostatic constant of proportionality, e is the charge of an electron, $\bar{c}_i$ is the mean thermal speed of ions, N_i is the concentration of ions, ε is the relative permittivity of the particle, and Z_i is the mobility of the ions.

In short, the diffusional charging process (Equation 4) is proportional to d_p whereas the field charging process (Equation 5) is proportional to d_p^2. The theoretical range of the particle-size dependent charging coefficient is therefore 1–

2, but since the particle charging is primarily dominated by the diffusion charging effect, the coefficient is typically closer to 1. The charging coefficient could be approximated using the Equations 4 and 5; however, it is difficult to accurately estimate, for instance, the concentration of ions in the proximity of particles, and, therefore, the charging coefficient and the charging efficiency is typically determined empirically for individual charger designs.
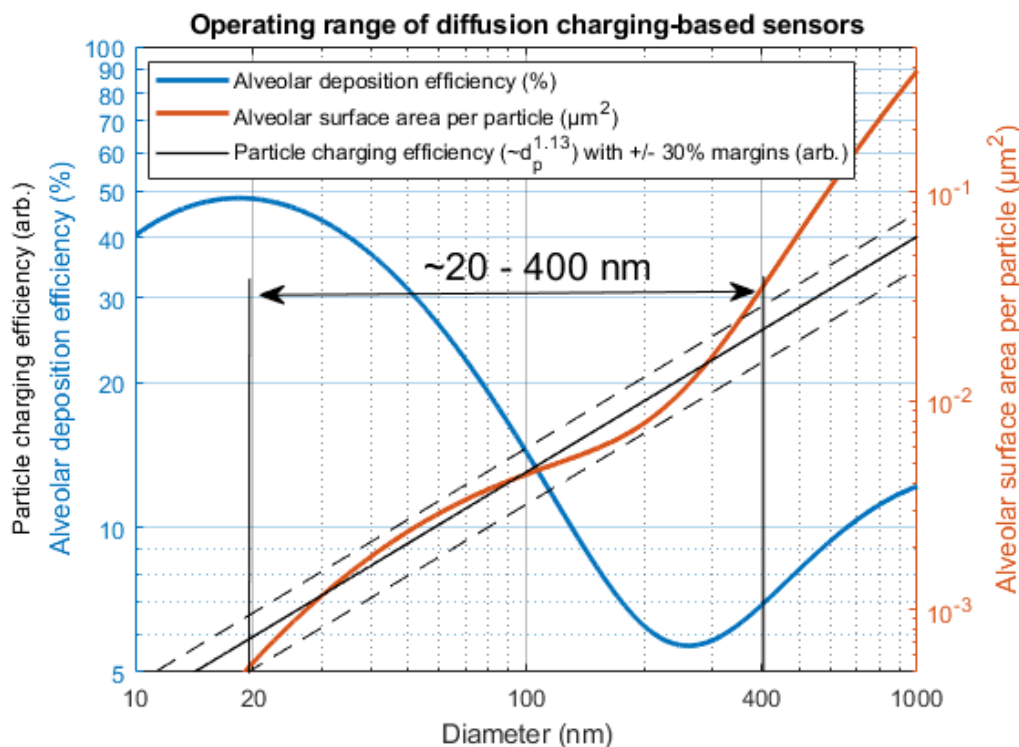
Figure 6. Operating range of a typical diffusion charging-based sensor (Todea et al. 2015).

The usefulness of a LDSA sensor to urban air quality measurements is highlighted by several different factors. As an alternative method, it bridges the gap between optical methods (> 300 nm) and ultrafine particles (< 100 nm), which is important considering the significance of ultrafine particles in urban air. Furthermore, the operating range of ~20 – 400 nm (Todea et al. 2017; Bau et al. 2015; Rostedt et al. 2014) implies that vehicular exhaust emissions and residential wood combustion are particularly well-suited for diffusion charging-type measurement (Brines et al. 2014; Lamberg et al. 2011; Tissari et al. 2008). The basic structure of a LDSA sensor is fairly simple, although not as simple as in optical sensors as the number of essential components is slightly higher. Nevertheless, it can be designed to incorporate the main features which characterize sensor type devices, such as small size (Rostedt and Keskinen 2018). Mixing of the sample particles and ions is typically carried out using an ejector pump which, when coupled with the non-collective characteristics of the escaping current measurement, makes the sensor to have relatively infrequent service interval. Currently, LDSA sensors are more expensive than optical sensors (e.g. ~5,000–20,000 versus 10–5,000 €), however, the price comparison is not entirely fair for several different reasons.

For the sake of clarity, some terminology and related practical differences between optical and diffusion charging-based sensors are explained here. In scientific literature, the term 'sensor' is often used loosely when referring to any type of sensor. However, to be precise, there is an important distinction to be made between a bare electronic component used to sense particles and a fully functioning sensor system. The bare electronic sensor component, which is produced by the Original Equipment Manufacturer (OEM), cannot be utilized in air quality measurements as such without additional data acquisition hardware and weather protective housing. A sensor system (also known as 'a sensor node'), on the other hand, is a turnkey measurement solution which, in addition to the OEM sensor module, includes, for instance, sampling system, data acquisition and user interfaces, and necessary weather protective housing. The distinction between 'sensor system' and 'an OEM sensor' is important as optical sensors are predominantly categorized as OEM sensors and diffusion charging-based sensors as fully commercialised sensor systems. Consequently, the unit price comparison between the two types is problematic. Besides the slightly different product segmentation, LDSA as an aerosol parameter is still relatively unfamiliar to the broader atmospheric community, and its measurement is not as common as PM2.5, for instance. This leads to smaller production volumes and increased per unit costs of sensors. An overview of the currently commercially available optical and diffusion charging-based sensors is shown in Table 1. The table is not entirely complete, and it is subject to change as the market evolves, however, it does include sensors most commonly appearing in the scientific literature. Additionally, the unit prices may be perceived as guidelines rather than absolute values. The optical sensors shown here are categorized as OEM sensors and the diffusion charging-based sensors as complete sensor systems. Sensor systems based on optical OEM sensors are also available with varying hardware configurations (from different companies), but most of the literature is based on the bare OEM sensor evaluations. Throughout this thesis, if not stated otherwise, the word 'sensor' is used synonymously for both OEM sensors and sensor systems.

Table 1. List of some of the commercially available optical and diffusion charging-based sensors.

Type	Manufacturer and origin	Model(s)	Price (€)
Optical	Alphasense, UK	OPC-N2, OPC-N3	500
	Dylos, USA	DC 1100 pro, DC 1700	300-400
	Honeywell, USA	HPMA115S0	35
	Nova fitness, China	SDS011	35
	Omron, Japan	B5W-ld0101	15
	Plantower, China	PMS 3003/5003/7003	20
	Sensirion, Switzerland	SPS30	40
	Sharp Corporation, Japan	GP2Y1010AU0F	10
	Shinyei, Japan	PPD42NS, PPD60PV	15-200
Diffusion charging	Dekati, Finland	DEPS	Undisclosed
	Naneos, Switzerland	Partector 1/2	6300-9000
	Pegasor, Finland	AQ Urban	Undisclosed
	Testo, Germany	DiSCmini	Undisclosed

2.5 Aerosol sensors in air quality monitoring

Although aerosol sensors, and optical aerosol sensors in particular, have been under intense investigation for some years now, there are no uniform standardization or regulation that addresses them. Consequently, there is no uniform definition to what constitutes a sensor. Current European Union Ambient Air Quality Directive 2008/EC/50 recognizes two different uncertainty levels for PM2.5 and PM10 measurement accuracy; continuous and indicative (25 % and 50 % relative uncertainty, respectively), however, it does not factor in any requirements regarding device hardware or software (e.g. physical size, unit price, data transfer) (European Council 2008). Respectively, air quality regulation set by the Environmental Protection Agency (EPA) of United States does not address sensors as an individual entity (Environmental Protection Agency 2007). Unofficially, a sensor is considered to be small or handheld, portable, and easy to use. Wireless data transfer is an integral part of a sensor and its unit price is supposed to be significantly lower than that of the standardized instruments (e.g. ~100 – 1,000 as opposed to ~10,000 – 100,000 €). Requirements for the measurement performance, such as accuracy and precision, are assumed to be modest, however, the sensor needs to be able to produce meaningful and consistent data. It is expected that official regulation addressing different aspects of air quality sensors (including gas sensors) will be implemented in the near future (Williams et al. 2019; F. Karagulian et al. 2019a). Currently, the lack of legislation and standardization limits large scale adoption of sensors to national, regional, and municipal air quality monitoring strategies.

Several articles evaluating the features and properties of different sensors has been published in the scientific literature. For the optical sensors, extensive review studies have been conducted by Karagulian et al. (2019b) and Rai et al. (2017), which show that, in general, optical sensors appear to exhibit fairly

promising performance; recorded coefficient of determination values (R^2) were typically greater than 0.50, when the sensor data was compared to a reference instrument (Figure 7, data adapted from Rai et al. 2017), and coefficient of variations (CV) were found to be in 1–28 % range. Coefficient of determination describes the proportion of variance (from 0 to 1, where 1 indicates no variance) for a dependent variable (i.e. sensor data) that is explained or predicted by the independent variable (i.e. reference data) in a regression model, and its mathematical formula is shown in Equation 9 (section 3.4). Coefficient of variation (also known as relative standard deviation, RSD) is a metric defined as the ratio of the standard deviation to the mean value, and, in this context, it has been used to indicate the precision of a sensor (i.e. the spread of measured values). Both review studies, however, point out that due to the multitude of used evaluation methods (e.g. both field and laboratory tests and variety of different regression models), it is difficult to draw a conclusive summary about the performance of optical sensors. Moreover, even though the R^2 is the most commonly reported metric, its incompleteness as a sole indicator for the sensor performance is acknowledged. Factors altering the response of optical sensors are reported to be, at least, high relative humidity and consequent hygroscopic growth of particles (e.g. Di Antonio et al. 2018; Jayaratne et al. 2018), particle composition and particle size (e.g. Levy Zamora et al. 2019; Wang et al. 2015; Northcross et al. 2013). Nonlinearity and saturation as well as long-term drift of the sensor response has also been reported (e.g. Li et al. 2019; Malings et al. 2019; Johnson et al. 2018; Austin et al. 2015). Due to these factors, it has been emphasized that the risk of sensor misuse is high and that if not addressed properly, optical sensors, when considering their current state of development, are unlikely to be fit for long-term regulatory monitoring. Nevertheless, their prospective nature is still underlined (Bulot et al. 2019; Liu et al. 2019; Sayahi et al. 2019).
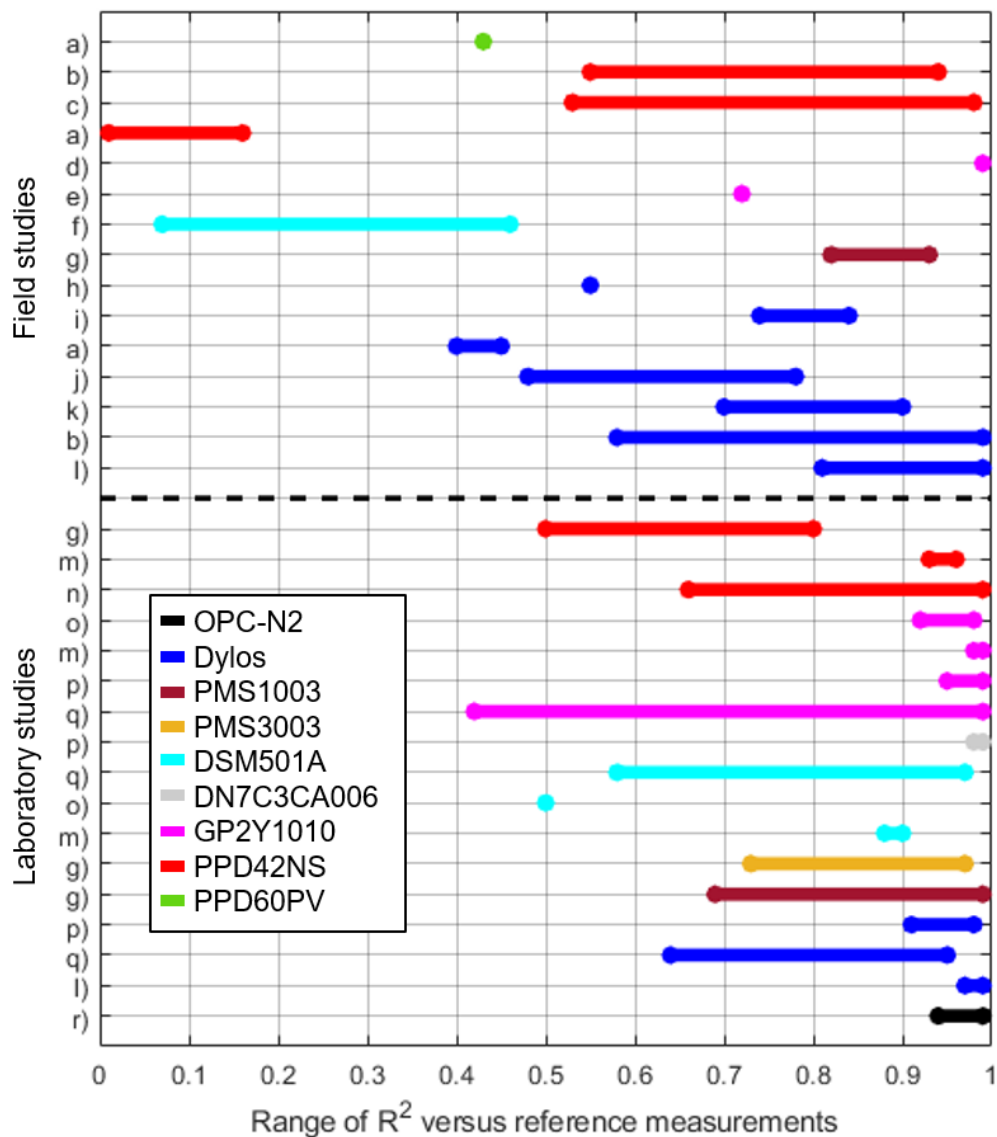
Figure 7. Summary of the R^2 values obtained from different sensor evaluation studies. The alphabets refer to following studies: a) Jiao et al. 2016, b) Holstius et al. 2014, c) Gao et al. 2015, d) Olivares et al. 2012, e) Olivares and Edwards 2015, f) Zikova et al. 2017, g) Kelly et al. 2017, h) Williams et al. 2014, i) Jovašević-Stojanović et al. 2015, j) Han et al. 2017, k) Steinle et al. 2015, l) Northcross et al. 2013, m) Wang et al. 2015, n) Austin et al. 2015, o) Alvarado et al. 2015, p) Sousan et al. 2016, q) Sousan et al. 2016a.

Apart from the Dekati DEPS, the accuracy and comparability of the diffusion charging-based sensors listed in Table 1 have been evaluated previously in multiple different laboratory experiments (e.g. Todea et al. 2015, 2017; Bau et al. 2015; Rostedt et al. 2014). Overall, it is concluded that, in most of the cases, an accuracy of ± 30 % can be expected for the measurement of LDSA if the particle size distribution is within 15 – 400 nm range. Different particle compositions and morphologies (excluding fibers) are reported to have little to no effect on the response, but it is noted that particles larger than 400 nm may lead to significant bias in the measured concentrations (Bau et al. 2012; Asbach et al. 2009). Hence use of a size-selective inlet is recommended. Diffusion charging-

based sensors have been used and tested in field measurement campaigns, although not as extensively as optical sensors (Dal Maso et al. 2016; Järvinen et al. 2015). Much of the research has also focused on the direct assessments of LDSA concentrations rather than the sensor evaluations themselves (e.g. Hama et al. 2017; Kiriya et al. 2017; Kuuluvainen et al. 2016; Eeftens et al. 2015; Reche et al. 2015). A review of measurement techniques for workplace nanoparticle exposures noted that while the level of accuracy of diffusion charging-based sensors is fairly good, they are not on par with the performance of conventionally used research instrumentation, such as the Scanning Mobility Particle Sizers (SMPS, TSI Inc., USA) or Differential Mobility Particle Sizers (DMPS) (Asbach et al. 2017). However, the relatively low cost, ease-of-use, and high mobility resulting from the small physical size are described to be definite advantages of the diffusion charging-based sensors.

Applications of optical sensor networks have been demonstrated, however, the total amount of these types of studies is sparse (Yuval et al. 2019; Masiol et al. 2018; English et al. 2017; Jiao et al. 2016; Gao et al. 2015). To the author's knowledge, no network studies have been conducted using diffusion charging-based sensors, apart from the Publication 4 presented in this thesis. The scarcity of these types of measurements is, at least partly, due to the substantial effort required to conduct such an experiment. For example, Caubel et al. (2019) conducted a sensor network study of 100 BC sensors for 100 days, and noted hardware and data acquisition redundancies to be very important factors to consider when designing the data quality assurance and control strategies. At the end of the experiment, 85 of the total 128 sensors (spare ones included) were operational, and repairs had to be carried out in the field throughout the experiment. Cellular reception was unavailable or unreliable in some parts of the neighborhood and thus manual data collection via SD cards was required. The authors said to benefit greatly from the help of a local community organization but carrying out the experiment was still described to be exceptionally challenging. Also, the sheer amount of data produced by the network translated into months-long analysis and validation work. But nevertheless, unprecedented data regarding the local variability of PM was obtained. Typical benefits of a network type data are reported to be, for instance, identification and characterization of pollution 'hotspots' (Gao et al. 2015) and addressing differences in temporal variations between different sites (Yuval et al. 2019). Additionally, it is worth noting that increased awareness and understanding regarding air pollution has been observed amongst the local communities which have participated in the experiments (English et al. 2017; Jiao et al. 2016).

When comparing the literature of optical and diffusion charging-based sensors, it appears that the diffusion charger-based sensors are more mature in terms of their technological development. The availability of commercial sensor systems, which have been designed for scientific research, as well as the more uniform methodology used to evaluate them, supports this. The recorded levels of accuracy are better than in most of the evaluations of optical sensors, and the specific response characteristics of the LDSA sensors have been assessed more comprehensively: for instance, the response characteristics of the Pegasor AQ

Urban, which is the main LDSA sensor used in this thesis (Publications 3 and 4), has been thoroughly evaluated from both theoretical and experimental point of view by Rostedt et al. (2014). To a large extent, similar research has not been conducted for optical sensors even though the sheer amount of studies focused on optical sensors is, nevertheless, greater. It is unclear why response characterizations for optical sensors have not been conducted.

3. Experimental methods

The experimental methods used in this thesis are divided into two separate entities; methods used in the response characterizations of optical sensors (section 3.3, Publications 1 and 2) and methods used in field evaluations (section 3.4, Publications 3 and 4). The instrumentation which was used in these studies, both reference devices and sensors, are described in sections 3.1 and 3.2, respectively, and the used data analysis methods in section 3.5.

3.1 Reference instruments

In Publications 1 and 2, reference values for the optical sensors were measured with Aerodynamic Particle Sizer (APS, TSI Inc., USA) and with GRIMM model 1.108 (GRIMM Aerosol Technik Ainring GmbH & Co. KG, Germany). The APS was used for primary characterization of sensor responses whereas the GRIMM was used to validate the obtained sensor response results (Publication 1) and demonstrate proper functionality of particle bin sizing (Publication 2). Working principle of the APS is predicated on time of flight of particles (aerodynamic diameters of 0.5 − 20 μm with 52 bins), and its performance has been thoroughly evaluated previously; it has a tendency to underestimate particle counts but particle sizing is comparable to that of a cascade impactor (Peters 2006; Peters and Leith 2003). The optical GRIMM is a mid-cost (10,000 − 15,000 €) aerosol spectrometer which measures particles from 0.23 to 20 μm with 15 size bins. Its response has been reported to being similar to the APS although some systematic particle size-dependent differences have been observed (Peters et al. 2006). Nevertheless, the accuracy of the GRIMM for mass measurements has been shown to being comparable to the standardized method of filter weighing (Burkart et al. 2010).

In Publications 3 and 4, optical and diffusion charging-based sensors were compared to Tapered element oscillating microbalance (TEOM 1405D, Thermo Fisher Scientific Inc., USA) and DMPS, respectively. TEOM is a reference grade instrument which adheres to the standardized uncertainty criteria of PM2.5 and PM10 measurements (European Council 2008). However, due to the filter weighing type measurement technique, volatile compounds evaporating from the filter make it susceptible to some inaccuracies (Allen et al. 1997). Reference values for the LDSA concentrations were measured with a DMPS which used

Vienna type differential mobility analyser (DMA) and Airmodus A20 condensation particle counter (CPC, Airmodus Oy, Finland). Measured particle size range was 6 – 800 nm (mobility diameter). LDSA concentrations were calculated from the DMPS data by weighing the size-resolved number concentrations with alveolar deposition fraction, and then calculating the total surface area accordingly. Particle size-dependent deposition fractions for different human respiratory tracts have been presented by the International Commission on Radiological Protection (ICRP) (Bair 1994). Albeit mobility particle size spectrometers, such as the DMPS, have not been standardized, they are commonly used in scientific research (Wiedensohler et al. 2012).

LDSA concentrations measured with the sensors were also compared to BC in Publication 3, and additionally, to NOx and PM2.5 in Publication 4. BC measurements were conducted with Multiangle Absorption Photometer model 5012 (MAAP, Thermo Fisher Scientific Inc., USA) in Publication 3, and additionally, with AE33 Aethalometer (Magee Scientific Co., USA) in Publication 4. In Publication 4, BC measurements of the MAAP and AE33 were unified by multiplying the AE33 values with 0.75, as suggested by Helin et al. (2018). NOx measurements were conducted with Horiba APNA models 360 and 370 (HORIBA Ltd., USA) and with Thermo 42i (Thermo Fisher Scientific Inc., USA). For PM2.5, measurements were conducted with TEOM, as described previously, and also with a beta-attenuation based FH 62 I-R monitor (Thermo Fisher Scientific Inc., USA). Different units were used in different stations NOx and PM2.5 measurements. In Publication 4, ancillary measurements for meteorological parameters were conducted with Vaisala's HMP155 (temperature and relative humidity) and WMT703 (wind speed and direction) monitors (Vaisala Oyj, Finland).

3.2   Evaluated aerosol sensors

The optical and diffusion charging-based sensors evaluated in this thesis, and their main properties, are listed in Tables 2 and 3, respectively. The sensors represent most of the ones evaluated in the literature, although, due to the continuous progress and development, new sensors are continuously introduced to markets and scientific literature. All the optical sensors evaluated here were in a form of OEM sensors, and implementation of additional hardware (e.g. data acquisition and sensor housing) is discussed in sections 3.3 and 3.4 where the specifics of the sampling configurations are addressed. The diffusion charging-based sensors were used as such as they already included all the essential features needed to conduct measurements.

The detection configurations of the evaluated optical sensors are arranged in either 90- or 120-degree scattering angle, and an infrared (IR) light emitting diode (LED) or a red coloured laser diode is used as a light source. This configuration is placed into an injection moulded plastic frame which is, furthermore, placed onto a printed circuit board (PCB) where the scattering light signal is filtered, amplified, and analysed. The total number of components in the setup is fairly low and the overall design is simple. The sensor output is either digital or analogue. Data from the digital sensors was obtained by reading sensor IC

registry, and data from the analogue sensors was obtained by counting the number of pulses (Omron B5W-ld01010), measuring the voltage level (Sharp GP2Y1010AU0F) and measuring a so-called Lo Pulse Occupancy (LPO) duration (Shinyei PPD42NS and PPD60PV). The LPO signal represents a ratio (0 – 100 %) of time in which the sensor is in a low voltage state. Data processing algorithms of the digital sensors were unavailable due to their proprietary nature, and in general, the availability of information regarding the electronic circuitry designs is limited. To the author's knowledge, only Sharp GP2Y1010AU0F and Shinyei PPD42NS sensors have been analysed for their electronic circuitry (Pedersini 2019; Canu et al. 2018).

The evaluated sensors were in original condition except the Shinyei PPD42NS and PPD60PV and Omron B5W-ld0101 sensors, which had their air heating resistors removed. The sample flow of these sensors was originally designed to be accomplished with natural convection resulting from the resistor-generated temperature gradient: however, this method of generating sample flow was considered to be too unreliable and inexact and hence, in both laboratory and field evaluations, the testing setup and assembled sensor systems included external vacuum pumps. External means of sample flow was utilized also for the other sensors (Publication 2), of which the PMS5003, SDS011, and SPS30 included built-in miniature fans and the GP2Y1010AU0F relied on plain diffusion. The reasoning behind the usage of additional vacuum pumps is described in detail in section 3.3. Sensors PPD42NS and PPD60PV were evaluated in Publications 1 and 3 whereas SDS011, B5W-ld0101, PMS5003, SPS30, and additionally PPD42NS, were evaluated in Publication 2.

Table 2. List of evaluated optical sensors and their main properties.

Sensor make and origin	Model(s)	Detectable size range*	Scattering angle and wavelength	Sensor output
Nova fitness, China	SDS011	0.3 – 10 µm	90°, Red (laser)	Digital; PM2.5, PM10
Omron, Japan	B5W-ld0101	> 0.5 µm	120°, IR (LED)	Analogue; pulse count
Plantower, China	PMS5003	0.3 – 10 µm	90°, Red (laser)	Digital; PM1, PM2.5, PM10
Sensirion, Switzerland	SPS30	0.3 – 10 µm	90°, Red (laser)	Digital; PM1, PM2.5, PM4, PM10
Sharp, Japan	GP2Y1010 AU0F	n/a	120°, IR (LED)	Analogue; voltage level
Shinyei, Japan	PPD42NS, PPD60PV	> 1.0 µm, > 0.5 µm	120°, IR (LED)	Analogue; PWM-signal

* According to the technical specifications sheet.

The Partector and DiSCmini are handheld diffusion charging sensors which measure LDSA concentrations using particle collection and a Faraday cup elec-

trometer type measurement. Their primary target application is personal exposure assessments, but they can be used in outdoor measurements with minor modifications (e.g. by implementing weather protection). In comparison, the Pegasor AQ Urban is a self-contained sensor specifically designed for outdoor operation. The main difference between these sensors is the sample pre-treatment system of the AQ Urban which dries the aerosol and prevents water condensation inside the sensor. Furthermore, the AQ Urban uses escaping current measurement technique which makes it to have a fairly infrequent service interval. The Partector and DiSCmini sensors require periodic cleaning due to dust accumulation in the Faraday cage filter. The diffusion charging-based sensors evaluated in this thesis were commercially available sensor systems and no additional modifications were done to them. DiSCmini and Partector sensors were placed inside the main measurement container and thus no additional weather protection was required. DiSCmini and Partector sensors were evaluated in Publication 3 and Pegasor AQ Urban in Publications 3 and 4.

Table 3. List of evaluated diffusion charging-based sensors and their main properties.

Sensor make and origin	Model	Detectable size range	Measurement type, primary application
Naneos, Switzerland	Partector 1	20 – 400 nm	Faraday cup electrometer, personal exposure
Pegasor, Finland	AQ Urban	10 – 400 nm	Escaping current, outdoor sensor
Testo, Germany	DiSCmini	10 – 700 nm*	Faraday cup electrometer, personal exposure

* Accurate size range is declared to be 10 – 300 nm.

3.3 Response characterization of optical sensors

Laboratory evaluation of the particle size-selectivity of optical sensors are addressed in Publications 1 and 2. In these publications, a novel method for producing reference aerosols with varying properties was developed and utilized. The key instrument used was a Vibrating Orifice Aerosol Generator (VOAG, TSI Inc., USA), whose working principle is predicated on the instability and breakup of a liquid jet column; mechanical vibrations of the orifice cause the protruding liquid jet column to disintegrate into uniform droplets, which evaporate and form particles from the non-volatile solute dissolved in the volatile liquid (Berglund and Liu 1973). If the liquid is non-volatile, the droplet diameter and particle diameter are equal. Otherwise, the formed particle size is calculable from the volumetric fraction of the non-volatile solute dissolved in the volatile liquid (Equations 6-7). The main deficiency of the VOAG (and the main limitation of the developed evaluation method) is that its smallest producible particle size is limited by the impurity within the carrier solvent and is in practice limited to approximately 0.5 μm.

$$d_d = \left(\frac{6Q}{\pi f}\right)^{1/3} \tag{6}$$

where d_d is the produced droplet diameter, Q is the liquid solution feed rate, and f is the vibration frequency.

$$d_p = (C + I)^{1/3} * d_d \tag{7}$$

where d_p is the resulting particle diameter, C is the volumetric concentration of the non-volatile solute dissolved in the volatile liquid, and I is the volumetric fraction of impurity within the volatile liquid.

The produced particle number concentration of the VOAG is dependent on the ratio of orifice vibration frequency and volumetric flow rate of the dilution air and, according Liu and Berglund (1973), a relative standard deviation (RSD) of less than 3 % is achievable if the aforesaid parameters are kept constant. Moreover, the formed particle size distribution is monodisperse having geometric standard deviation (GSD) as low as 1.014 (Berglund and Liu 1973). These features, along with ability to calculate the particle size from the liquid concentrations, allows the VOAG to output primary standard aerosols.

The developed evaluation method used in this thesis is based on the notion that blending of two liquid solutions with different non-volatile concentrations produces a stable particle size gradient respective of the concentrations of the blended solutions. In other words, the produced particle size can be controlled by controlling the concentrations of the blending solutions, and the running parameters of the VOAG can be kept constant throughout the experiment. This enables continuous production of different sized particles and higher experiment reproducibility as the need to manually alter and tune different solutions and VOAG running parameters is eliminated. Furthermore, rather than settling for a fixed amount of different particle sizes or test points, the gradient-like output allows, in theory, investigation of indefinite test points. An example of the produced particle size gradient is shown in Figure 8.
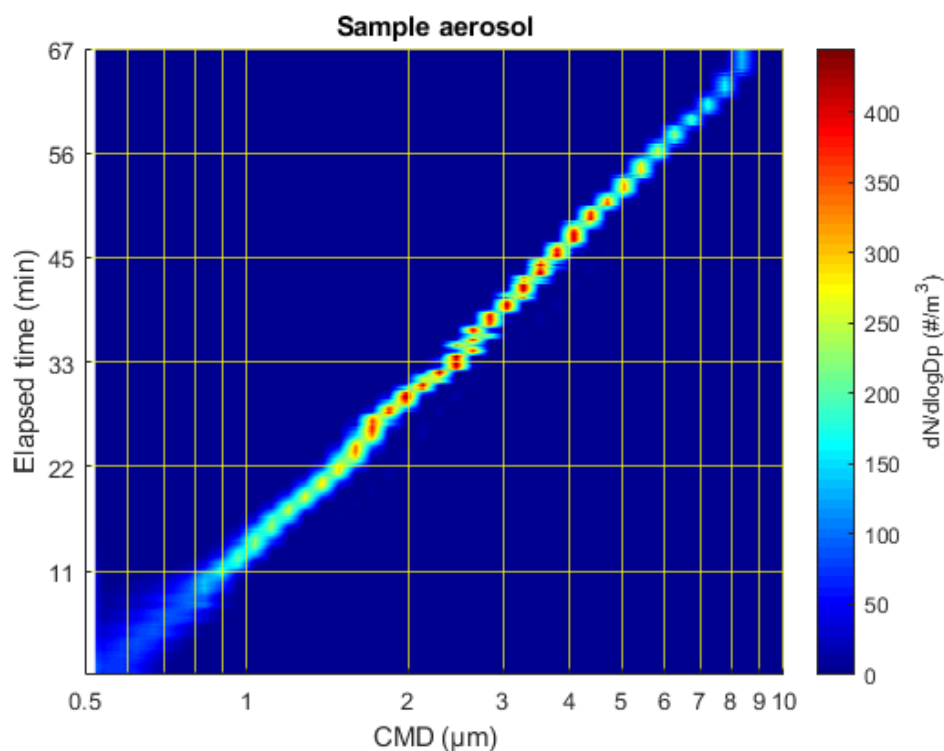
Figure 8. An example of the reference particle size gradient (adapted from Publication 2). The decreasing number concentrations in particle sizes below 1 μm and above 5 μm result from the decreasing detection efficiency of the APS and increasing inertial deposition losses in the sampling lines, respectively.

In Publication 1, the blending of different solutions was achieved by using two separate syringe pumps and a manually operated three-way valve. A test run began by initiating the liquid jet column of the VOAG with a pure 2-propanol (> 99.999 %, Sigma-Aldrich). Here, the formed particles resulted from the impurities within 2-propanol (1 ppm, approximately 0.5 μm sized particles). After a steady state was achieved, the three-way valve was switched to feed liquid from the parallel syringe pump, which was operated with either of the two aerosol solutions; dioctyl sebacate-2-propanol (DOS) or palmitic acid-2-propanol solution (PA) (concentrations of 10 g L^{-1} in 2-propanol). Experiments were done with both solutions as this allowed investigation whether the particle composition influenced the sensor responses. Particles resulting from the DOS were transparent oil droplets whereas particles resulting from the PA were white crystalline particles. The blending of the pure 2-propanol and aerosol solution (DOS or PA) resulted in a monodisperse particle size gradient (approximately from 0.5 to 10 μm) over a ~ 12-minute time period. The produced reference aerosol was divided to the APS (reference instrument) and evaluated sensor (Shinyei PPD42NS or PPD60PV) with a Y-branch, and the sensor response was compared to that of the APS. Data resolution of both APS and sensor was 10 seconds. After the laboratory experiments were conducted, the obtained results were validated in an 18-day field campaign. The campaign was conducted at the SMEARIII station in Helsinki, Finland (urban background, see e.g. Järvi et al. 2009), and a GRIMM 1.108 aerosol spectrometer was used as a reference instrument. The validation was done by comparing the sensor output to the mass

fraction of the GRIMM which was supposed to correspond the detectable particle size range of the sensor. Data resolution in the field validation was 1-hour average.

In both laboratory experiments and field validations, the OEM Shinyei sensors were housed in an airtight aluminium alloy enclosure which was equipped with a miniature vacuum pump (sample flow of 0.9 L min^{-1}). Both sensors were placed in the same enclosure as preliminary examination had shown that the different models had different response characteristics. This way, when the complete sensor system (hereafter referred as Prototype aerosol sensor, PAS) was deployed to the field (field validation of Publication 1, used also in Publication 3), two different outputs could be obtained for two different particle size fractions, and the obtained results were to be representative to that of the laboratory evaluation. In the enclosure, the Shinyei sensors were positioned in a series configuration (one after each other) so that a common inlet line could be used. The connection between the sensors was implemented in a way that sampling losses were minimised, and a stable sample flow was passing through both sensors. In general, the plastic body layouts of optical sensors have not been optimised for ambient sampling and it is not uncommon that this issue is overlooked even in scientific studies. Many of the available sensors has 90-degree bends incorporated in their plastic layouts, which poses a risk of particle inertial deposition. Furthermore, the plastic bodies themselves are statically charged, which may entail additional electrostatic losses.

Although Publication 1 showed that the concept of the developed laboratory evaluation method was valid, there were few shortcomings and places of improvement which were addressed in Publication 2. First and foremost, while the blending of the two solutions using separate syringes and a manual three-way valve was found to be feasible, the rate at which the particle size gradient was evolving could not be controlled. Consequently, a single test run lasted less than 15 minutes and the amount of obtained data points was small. The quickness of a test run can be perceived advantageous, particularly when considering preliminary type of testing, however, stronger statistical power and scientific robustness were considered to be more important in this case. Another important weakness of the syringe pump setup was the fact that manually switching the feed from one syringe to another sometimes caused a temporary pressure spike in the liquid. This, in turn, resulted in instability in the VOAG output and, although the operation of the VOAG was already made more straightforward, there was no way to avoid this without substantial re-configuration of the liquid feeding system.

In Publication 2, the syringe pump feeding was replaced with a GP50 gradient elution pump (Dionex GP50, Thermo Fisher Scientific Inc., USA). The GP50 was originally designed to be used ion-chromatography and it includes several important features, which make it well-suited for the application at hand. The GP50 has four different eluent channels (liquid reservoirs) and it is capable of dispensing liquids with high pressure (max. 5000 psi) and precise volume flow rate (0.04 − 10.0 mL min^{-1} in increments of 0.01 mL min^{-1}). The four eluent channels can be mixed together with a resolution of 0.1 % (combined output of

the four channels always 100 %), and, moreover, the GP50 has a user-interface which enables the operator to generate parameterized eluent dispensing programs. Unlike previously, utilization of the GP50 allows the user to control the rate at which the particle size is changing, and the blending of the solutions is conducted automatically by the pump itself. Essentially, the GP50 allows the user to freely choose and produce monodisperse aerosols of desired particle sizes in a fully automated manner.

Six different sensor models were evaluated in Publication 2 (Table 2, all sensors except Shinyei PPD60PV), and the internal precision of these models was assessed by repeating the tests for three individual units. The tests were conducted with a DOS solution only. The GP50 was operated with a 10-step dispensing program, which produced monodisperse aerosols from ~ 0.5 to 8.5 μm over a 70-minute time period. Data from the APS and sensor was acquired synchronously in 10-second resolution. The used sampling configuration was the same as in Publication 1 except for the Y-branch which was replaced with an isokinetic flow splitter. Previously, the sample flow was divided unevenly to the APS and sensor as the respective flow rates were 5 and 0.9 L min⁻¹. Here, the sample flow was evened out by connecting the aerosol flow of the APS (1 L min⁻¹) directly to the flow splitter and replacing the miniature vacuum pump of the sensor enclosure with an external medium sized vacuum pump (flow rate adjusted to 1 L min⁻¹). The total flow of the APS (5 L min⁻¹) is a combination of aerosol flow (1 L min⁻¹) and sheath flow (4 L min⁻¹, taken from the laboratory air in this case). This setup ensured that the flow rate and particle size-dependent deposition losses were identical for both the APS and evaluated sensor. Also, instead of using the PAS or the aluminium alloy enclosure, as in Publication 2, the sensors were housed individually in custom designed (3d-printed) enclosures, which were optimised for minimal sampling losses (e.g. proper orientation of the sensor and an antistatic 3d-filament, see supplementary material of Publication 2). In principle, for some sensors it would have been possible to drill a hole to the plastic body and attach the sample line directly towards the optical detection chamber. However, this kind of configuration was believed to reflect poorly the real conditions in which the sensors are typically used, and thus, it was decided not to be done. A schematic of the final evaluation setup is shown in Figure 9.

To account for the possible discrepancy between the used 1 L min⁻¹ flow rate and the flow rate for which the sensors were originally designed for (i.e. convection-induced flow, built-in fans, and a plain diffusion), additional tests were conducted with flow rates of 0.5 and 2 L min⁻¹. The results confirmed that the ancillary flow rate did not distort the results (see supplementary material of Publication 2). This was expected as, theoretically, the sample flow rate of an optical particle counter is directly proportional to the frequency and duration of observed light pulses (i.e. particle number concentration), but not to the height of the pulses (i.e. particle size). As this research focused solely on the investigation of particle size discrimination characteristics of sensors, it is irrelevant whether the measured absolute concentrations were different from the true values. Moreover, photometer-type sensors do not have any means to discriminate

sizes of individual particles, and their measured concentration is not a function of sample flow rate. When considering the field studies conducted in Publications 1 and 3, it is possible that the flow rate of 0.9 L min^{-1}, which was the flow rate used with PAS, yielded unique slopes and intercepts for linear regression (assuming that the Shinyei sensors operated as particle counters). However, it is not evident how the sampling of a temperature gradient-based OEM sensor (i.e. the Shinyei PPD42NS and PPD60PV sensors with heating resistors) should be accomplished: it is highly probable that the resistor-generated flow rate is weak (i.e. small pressure differential) and thus it is also susceptible to dynamic effects of wind, for instance. This would imply that protective measures would have to be taken, which would, in turn, compromise the stability and representativeness of the sample (i.e. no constant sample forced through the sensor). To allow more even inter-comparison of different studies, it would be beneficial for the scientific community to establish a clear framework describing how the sensor evaluations should be conducted for different types of sensors. Such framework could include, for instance, computational fluid dynamic analysis (CFD) of the flow characteristics (laminar or turbulent flow) and flow rate-dependent particle losses (see e.g. Mongelluzzo et al. 2019) among other sampling related details.
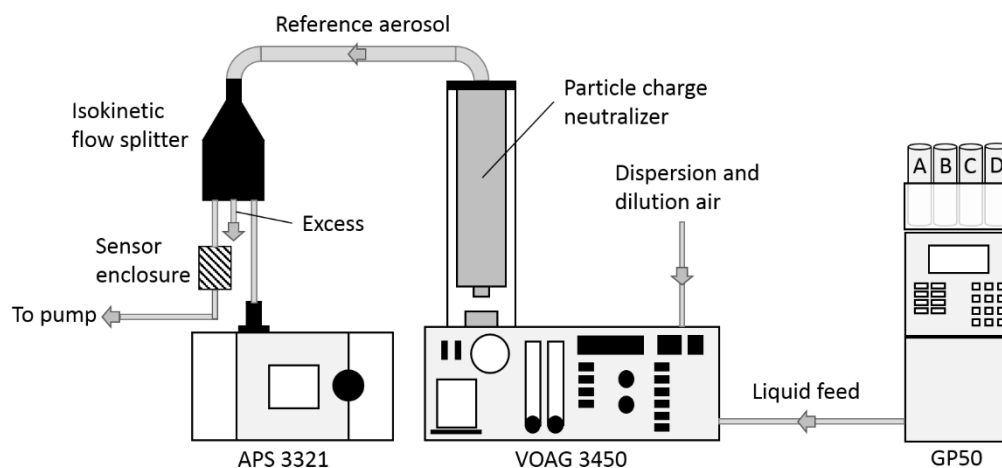
Figure 9. Evaluation setup used in Publication 2 (adapted from Publication 2). The symbols "ABCD" on the GP50 represent different eluent channels.

The particle size-selectivity analysis was conducted by comparing the raw sensor output signal to the total mass measured by the APS. Any measurement point which had GSD (calculated from the APS size distribution data) exceeding 1.2 was excluded from the analysis, but typically the GSD values were within 1.04 − 1.08 range. The sensor response was normalized using Equation 8 in order to prevent arbitrary unit comparisons and to make the cross comparison of different sensor models more meaningful.

$$Normalized\ detection\ efficiency = \frac{\frac{R_{S,i}}{R_{APS,i}}}{\max\left(\frac{R_S}{R_{APS}}\right)} \tag{8}$$

Where i is the ith measurement point, R_s is the sensor response signal, and R_{APS} is the APS total mass concentration.

Because the normalized 10-second resolution raw data was in a gradient-like form (no static test points for different particle sizes), it was divided into 30 logarithmically distributed size bins (from 0.45 to 9.73 μm) according to the count median diameters (CMD, aerodynamic) measured by the APS. For each of these size bins, an average sensor response was calculated as a function of average CMD. The decision to divide the data into 30 bins was based on the clarity of the produced figures and statistically sufficient number of measurement points belonging to each bin. This process was completed for three different sensor units, and a combined (average and standard deviation) sensor response was calculated. The valid detection range, which describes the particle size range the sensor is capable of measuring of, was defined as the upper half of the normalized detection efficiency curve. The sensor outputs were treated as discrete so that no overlapping signals were obtained. For instance, sensors having outputs PM1 and PM2.5 were used as PM1 and PM2.5-PM1 (size range of 1.0 − 2.5 μm).

3.4   Short and long-term field measurement campaigns

Both short and long-term field measurement campaigns were carried out in the Helsinki metropolitan which is situated in the coastal area of southern Finland. The metropolitan area consists of four cities (Helsinki, Espoo, Vantaa, and Kauniainen) and approximately 500,000 citizens. The main sources of submicron PM are residential wood combustion, vehicular exhaust emissions from traffic, long-range transport, and secondary aerosol formation (Pirjola et al. 2017; Carbone et al. 2014; Timonen et al. 2013; Niemi et al. 2009; Saarikoski et al. 2008). Main source of coarse mode particles is street dust resulting from traction sanding and pavement wear (Kupiainen et al. 2005, 2016) and sea salt.

The short measurement campaign (Publication 3) was conducted at Mäkelänkatu Supersite station, Helsinki. The Supersite station is one of the fixed measurement sites of the Helsinki metropolitan area and it is operated and maintained by the Helsinki Region Environmental Services Authority (HSY). The station is located in a busy avenue street canyon right next to the street and it is heavily affected by traffic related aerosols; both combustion emitted nanoparticles and coarse mode street dust particles (Hietikko et al. 2018; Kupiainen et al. 2016). The traffic volume at Mäkelänkatu was 28,000 vehicles per workday in 2016 (statistics from the City of Helsinki) and the proportion of heavy-duty vehicles was 11 %. The measurement campaign lasted approximately 3 months, from November 15th 2016 to February 15th 2017. During this time, the optical sensor (PAS, Shinyei PPD42NS and PPV60PV sensors) and Pegasor AQ Urban were operated for the full duration of the campaign whereas the Partector and DiSCmini sensors were operated only ~ 1 week, from 16th to 23rd of November and from 18th to 23rd of November 2016, respectively. The PAS was essentially in the same form as in Publication 1; only the resolution of data acquisition was changed from 10-second to 1-hour average. The PAS and the handheld Partector

and DiSCmini sensors were placed inside the measurement container alongside with the reference instrumentation (TEOM and DMPS). The Pegasor AQ Urban was placed on top of the container roof. Inlets of all the sensors and reference instruments were approximately on the same height.

The long-term measurement campaign (Publication 4) lasted approximately one full year, from February 1st 2017 to 31st January 2018, and only Pegasor AQ Urban sensors were used. The measurements were conducted at four different sites which represented three distinctive environments found in the Helsinki metropolitan urban area. The Supersite station, where the measurements of Publication 3 were also conducted, was used to represent traffic environment. The other two environments were urban background site at Kallio, Helsinki and the detached housing site at Rekola, Vantaa.

The Kallio station (hereafter referred as urban background station, UB) is an urban background station situated near a sports field approximately 1.5km from Helsinki downtown. Distance to nearest busy roads are 80 m (Helsinginkatu, 5,000 vehicles per workday) and 300 m (Sturenkatu, 25,800 vehicles per workday), and lower PM concentrations are typically observed at Kallio than in the vicinity of busy roads. Approximately 320 m north of the station is a coffee roastery which periodically produces organic particles of ~ 100 nm in size (Carbone et al. 2014). Distance between the Supersite and UB stations is less than 1 km.

Rekola stations (hereafter referred as detached housing area 1 and 2; DH1 and DH2) are situated in a small house area in Vantaa, Finland. The distance between the stations is approximately 670 m. The sites are surrounded by detached houses, and air quality in Rekola is known to be strongly affected by emissions from residential wood combustion (Helin et al. 2018).

Supersite, UB and DH1 are fully equipped air quality stations with aerosol (PM2.5, PM10, black carbon) and trace gas (NOx, O3) measurements, whereas the DH2 station is a sensor site with only Pegasor AQ Urban sensor. The DH1 station is therefore used to represent detached housing area when the analysis of the observed differences between urban, urban background, and detached housing area environments are made. The meteorological data were obtained from Pasila mast (78 m above sea level). These data were considered to represent the general meteorological conditions in the metropolitan area.

In both short and long-term campaigns, the accuracy of the sensors was evaluated by using linear regression and corresponding correlation metrics of coefficient of determination (R^2), root-mean-square error (RMSE), and normalized root-mean-square error (NRMSE) (Equations 9-11). Although linear regression may not have been the optimal model in all the cases, its use is justified by the fact that it is the standard method used in official equivalency testing of a candidate and reference method (United States Environmental Protection Agency 2013; European Comission 2010).

$$R^2 = 1 - \frac{SSR}{SST} \qquad (9)$$

Where SSR is the sum of squared residuals with respect to the linear regression and SST is the sum of the total squared residuals with respect to the average value of the sensor.

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(x_i - y_{i,cal})^2}{n}} \tag{10}$$

$$NRMSE = \frac{RMSE}{\bar{x}} \tag{11}$$

Where n is the number of sample pairs, x_i is the ith sample of the reference value, $y_{i,cal}$ is the ith sample of the calibrated sensor value, and $\bar{x}$ is the mean of the reference values. Calibrated sensor values were used to prevent arbitrary unit comparisons (e.g., Lo Pulse Occupancy and µg m^{-3}, pA and µm^2 cm^{-3}).

4. Results and discussion

4.1   Particle size-selectivity of optical aerosol sensors

The size-selective response functions of the evaluated optical sensors are shown in Figure 10 a-f (Publication 2), and the complete list of valid detection ranges (including the results from Publication 1) are shown in Table 4. In Figure 10, the coloured circles represent the average responses calculated from the three tested units and the shaded background areas respective standard deviations. An exception to this is the Shinyei PPD42NS (Figure 10e) in which the responses of the three units are shown separately. The size fractions (size bins) shown in the figure legends are those declared by the manufacturers.

Overall, the results of the laboratory evaluations show that optical sensors exhibit widely different characteristics regarding their particle size-selectivity; from < 0.7 μm (bin 1 of Plantower PMS5003) to > 5.9 μm (Shinyei PPD42NS unit 3, Table 4). However, to be precise, none of the sensors adhered to the detection ranges declared by the manufacturers. For example, the SPS30 sensor (Figure 10c) is stated to have four different outputs for particle sizes of < 1, 1–2.5, 2.5–4.0, and 4.0–10 μm. From these outputs only the first bin 1 (< 0.9 μm) appears to be sufficiently close to the detection range declared in the specifications sheet (< 1.0 μm, PM1). The other three bins are significantly off of their stated detection ranges, and moreover, appear to exhibit nearly identical size-selectivity. This implies that the manufacturer may have calibrated the sensor using only two different type test aerosols, and thus, has obtained responses for only two different particle sizes ranges (bin 1 < 0.9 μm and bins 2, 3, and 4 0.7 − 1.3 μm). When considering field measurements, it is evident that varying results will be obtained; a preliminary study by South Coast Air Quality Management District (SCAQMD) showed that the correlation (R^2) between the SPS30 and a reference-grade GRIMM instrument decreased from 0.91 to 0.83 and further down to 0.12 when the sensor was evaluated for the accuracy of PM1, PM2.5, and PM10 measurements, respectively (SCAQMD 2019). The results are in line with observations of this thesis and illustrate how a sensor with limited operational range may exhibit a near regulatory grade accuracy when the measured size fraction is in align with its valid detection range. On the contrary, the severity of data misinterpretation becomes apparent when the sensor measurement is extended to cover particle sizes which it cannot measure (PM10).
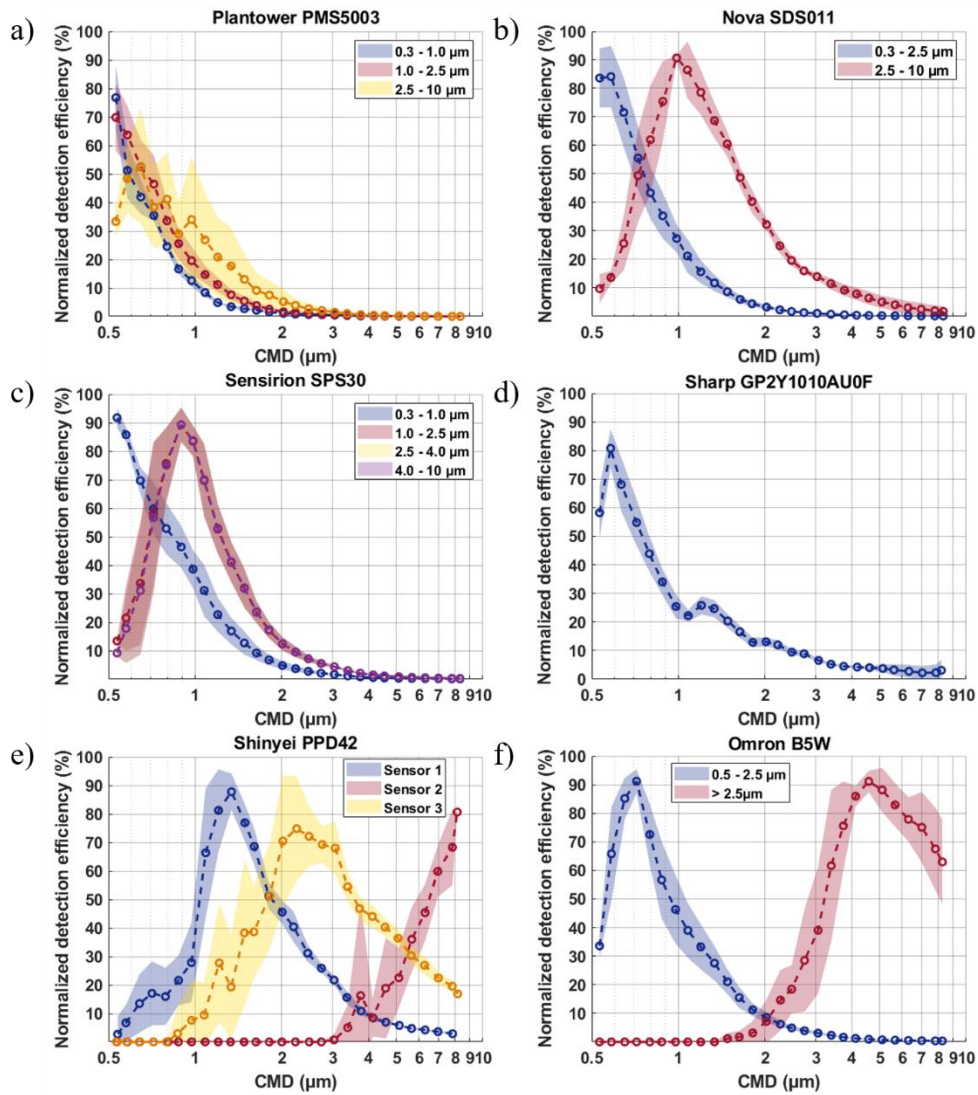
Figure 10. Particle size-selectivity of the optical sensors tested in Publication 2 (adapted from Publication 2).

Table 4. List of evaluated optical sensors and their valid detection ranges. Symbols "smaller than" and "greater than" refer to cases where the lower or upper cut-point of the detection range was beyond the size range of the produced reference aerosol. The detection ranges stated in the technical specification sheets of sensors are shown in parenthesis. Units in µm.

Sensor	Bin 1	Bin 2	Bin 3	Bin 4
Plantower PMS5003	< 0.7 (0.3-1.0)	< 0.8 (1.0-2.5)	< 1.0 (2.5-10)	-
Nova SDS011	< 0.8 (0.3-2.5)	$0.7 - 1.7$ (2.5-10)	-	-
Sensirion SPS30	< 0.9 (0.3-1.0)	$0.7 - 1.3$ (1.0-2.5)	$0.7 - 1.3$ (2.5-4.0)	$0.7 - 1.3$ (4.0-10)
Sharp GP2Y1010AU0F	< 0.8 (n/a)	-	-	-
Shinyei PPD42NS	$1.0 - 2.1^*$ (> 1.0)	$1.5 - 4.9^*$	$> 5.9^*$	$1.6 - 5.0^{**}$
Shinyei PPD60PV	$0.5 - 1.6^{**}$ (> 0.5)	-	-	-
Omron B5W-ld0101	$0.6 - 1.0$ (0.5-2.5)	> 3.2 (>2.5)	-	-

* Valid detection ranges of the individual OEM sensors and not bins.

** Result of Publication 1.

Results of the 18-day field validation experiment (Publication 1), where the Shinyei PPD42NS and PPV60PV sensors were tested, show similar results to that of the SPS30; the sensors achieve high measurement accuracies (R^2: 0.96–0.99) when the investigated particle size range is targeted to match the valid detection range of the sensor (Figure 11 a-d). On the other hand, studies, where the PPD42NS was used to measure PM2.5, have reported more modest R^2 values of 0.75 and 0.55 – 0.60 (Bai et al. 2019; Holstius et al. 2014) and even values as low as 0 – 0.28 (K. K. Johnson et al. 2018; N. E. Johnson et al. 2018). The differences in measured accuracies are most likely due to the changes in ambient particle size distribution and its comparability to the valid detection range of the sensor. Besides the Shinyei and SPS30 sensors, other sensors have exhibited similar characteristics (both high and low accuracy depending on the conditions); Plantower PMS5003 (e.g. Feenstra et al. 2019; Levy Zamora et al. 2019; Li et al. 2019; Magi et al. 2019; Malings et al. 2019; Laquai 2017), Nova SDS011 (e.g. Liu et al. 2019; Badura et al. 2018; Budde et al. 2018; Laquai 2017b), Sharp GP2Y1010AU0F (e.g. Li and Biswas 2017; Sousan et al. 2016; Alvarado et al. 2015; Wang et al. 2015; Budde et al. 2012).
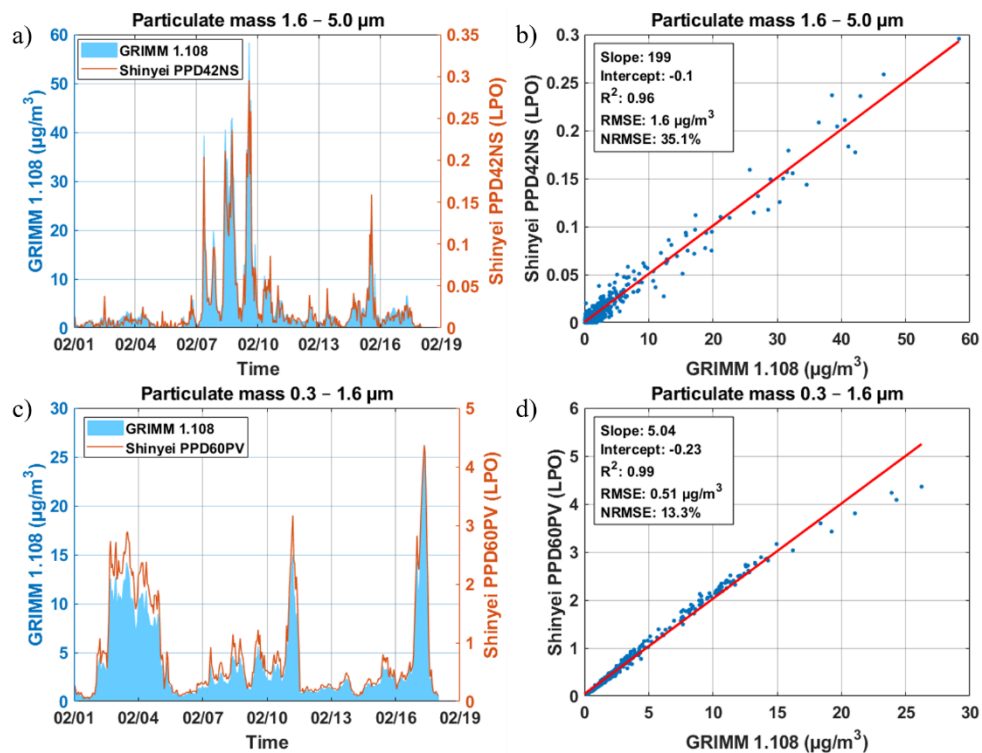
Figure 11. Field validation results (time series and respective correlation plots) of the Shinyei PPD42NS and PPD60PV sensors and GRIMM 1.108. Panels a-b correspond to particulate mass in size range 1.6 – 5.0 μm and panels c-d to 0.3 – 1.6 μm size range.

4.2 Applicability of sensors to urban air quality measurements

Correlation plots and respective regression metrics of the mass-based PM2.5 and PM2.5-10 measurements (PAS sensor system and TEOM) and LDSA (Pegasor AQ Urban, Partector, DiSCmini, and DMPS) are shown in Figure 12 a-d. The PAS, Partector, and DiSCmini data shown here was measured in the

three-month long campaign (Publication 3) and the Pegasor AQ data in the longer one-year campaign (Publication 4). Both measurements were made at the Supersite station.

The PAS sensor system (Shinyei PPD42NS and PPD60PV) shows worse accuracy when compared to the results of the validation test conducted in Publication 1 (Figure 11); R^2 values decrease from $0.96 - 0.99$ to $0.87 - 0.77$, and RMSE and NRMSE values from $1.6 - 0.51$ µg m^{-3} and $35.1 - 13.3$ % to $11 - 3.4$ µg m^{-3} and $78.5 - 40.9$ %, respectively. This is, evidently, due to the mismatch between the sensor response characteristics ($0.5 - 1.6$ and $1.6 - 5.0$ µm) and measured size fractions ($0 - 2.5$ and $2.5 - 10$ µm). The diffusion charging-based sensors (Pegasor AQ Urban, Partector, and DiSCmini), on the other hand, show higher accuracy (R^2: $0.90 - 0.97$) which is reasonable considering that the literature indicates them to be more mature in terms of their technological development.
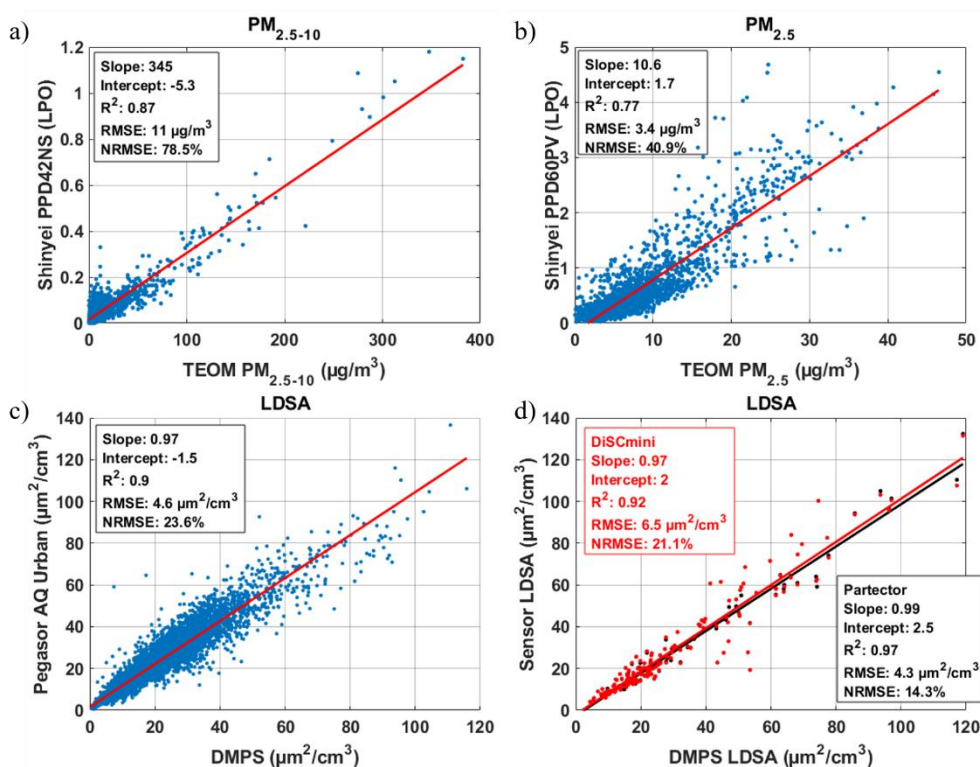

Figure 12. Summary of the correlations measured for the PAS and TEOM (a-b) and Pegasor AQ Urban (c), and Partector and DiSCmini (d), and DMPS.

The applicability of an optical sensor to urban air quality measurements is dictate by the sensor's valid detection range. Considering the level of accuracy of the PAS (Figure 12 a-b), it is debatable whether it is sufficient for long-term regulatory monitoring. As the response characteristics of the PAS and the standardized reference size fractions are not the same, the accuracy of the obtained results will depend on the chosen calibration factors (slope and intercept) and their representativeness regarding specific ambient size distributions. If the size distribution changes significantly, the disproportional weighing of different sized particles will lead to lower levels of accuracy. The calibration factors of the PAS could be determined so that typical episodic periods, such as the ones caused by street dust or long-range transported aerosols, could be measured as

accurately as possible; lower tail end of the size distribution of the street dust and long-range transported aerosols, which have had enough time to grow and form accumulation mode particles, are close to the optimal detection range of the PAS. However, this would naturally compromise the sensor accuracy when such episodes are not occurring. These trade-offs underline the need of the user to properly acknowledge the limitations and, moreover, to carefully consider how to ensure the validity and representativeness of the measured data.

To increase the accuracy of optical sensors, there is a growing number of studies successfully demonstrating the use of machine learning/artificial intelligence methods for the calibration of sensors (e.g. Concas et al. 2019; Borrego et al. 2018; Zimmerman et al. 2018). For instance, Si et al. (2019) showed that the use of a neural network-based calibration method improved the sensor accuracy considerably (Pearson's r from 0.74 to 0.85 and RMSE from (Si et al. 2019; Borrego et al. 2018) 9.93 to 3.91 µg m^{-3}), and that after the calibration, the variances of the measured PM2.5 values were no longer statistically significantly different from the PM2.5 variances measured with the reference instrument. Similar result could not be achieved using a simple linear regression. The weakness of advanced calibration methods is their lack transparency from a mathematical point of view (i.e. black-box algorithms) and their poor transferability between different sensor models and environments; results obtained from one study may not be directly reproducible by another study. Machine learning methods also require a dedicated training period in which the sensor is not producing calibrated data. This slows down the rate at which sensors can be deployed, although, it can be said that similar limitation applies for the use of simple linear regression as well.

Regarding combustion emitted nanoparticles, the PAS is poorly suited for their measurement. Although some portion of the traffic originated particles can be observed (Figure 13a), the diffusion charging-based approach achieves much higher sensitivity. In Figure 13a, the hourly mean values of LDSA and PM2.5, measured with Pegasor AQ Urban and PAS (Shinyei PPD60PV), have been normalized to represent the variances of measured concentrations occurring during a day (Publication 3). The normalization was done by calculating the average daily (24-hour average) value and then dividing it with the calculated hourly mean values. The shape of the LDSA cycle variation resembles closely to that of the traffic volume profile in which the morning and afternoon rush hours usually take place at 8–9 am and 4–5 pm, respectively (see Hietikko et al. 2018). The measured standard deviations of the normalized hourly mean concentrations of LDSA and PM2.5 are 0.37 and 0.07, respectively. Related to this, the measured BC is almost exclusively resulting from the adjacent traffic (incomplete combustion, Helin et al. 2018), and it exhibits relatively high correlation with LDSA (R^2 = 0.72, Figure 13b). The respective correlation for the PM2.5 measured with PAS is only 0.08. The high correlation between LDSA and BC is a common observation found in urban traffic sites (Hama et al. 2017; Eeftens et al. 2016; Reche et al. 2015), and it highlights the suitability of diffusion charging-based detection technique to the measurement of combustion emitted par-

ticles. This is an important observation considering that urban air quality is often affected most heavily by the vehicular exhaust emissions and residential wood combustion.

The distinctiveness of LDSA and its difference to PM2.5 is illustrated in Figure 14. The colour of the data points represents the particle mass median diameter (MMD) calculated from the size distribution data of DMPS, and it can be seen that larger MMD is linked to higher PM2.5 concentrations and vice versa. This is expected as particulate mass is proportional to the cube of particle diameter. However, for the LDSA, larger MMD does not directly translate into higher concentrations. This is because the particle alveolar deposition efficiency decreases significantly in the size range of 20 – 300 nm (from ~ 50 to 6 %), and furthermore, because of the surface area of particles is proportional only to the square of particle diameter. In larger particle size fractions (e.g. > 500 nm) the total number of particles is also significantly lower. Thus, LDSA should not be treated as a simple extension of PM2.5, for example, but rather as an unique and independent air quality parameter.
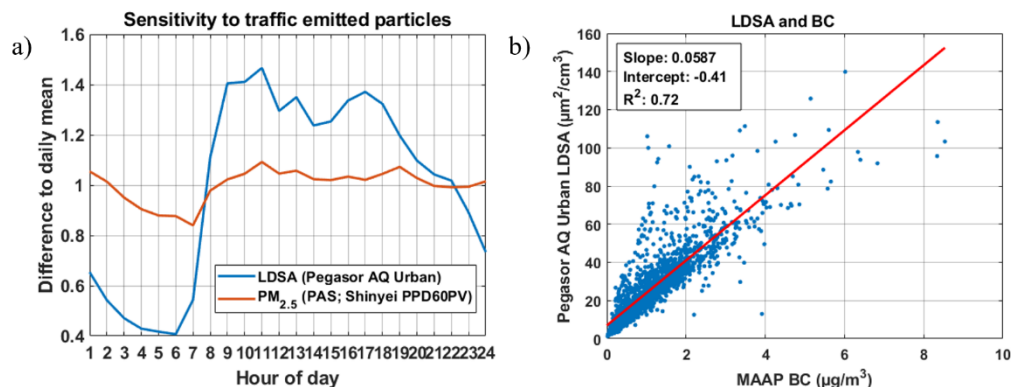
Figure 13. Normalized sensitivity of the Pegasor AQ Urban and PAS to traffic emitted particles (a) and correlation of the LDSA and BC (b) measured with the Pegasor AQ Urban and MAAP.
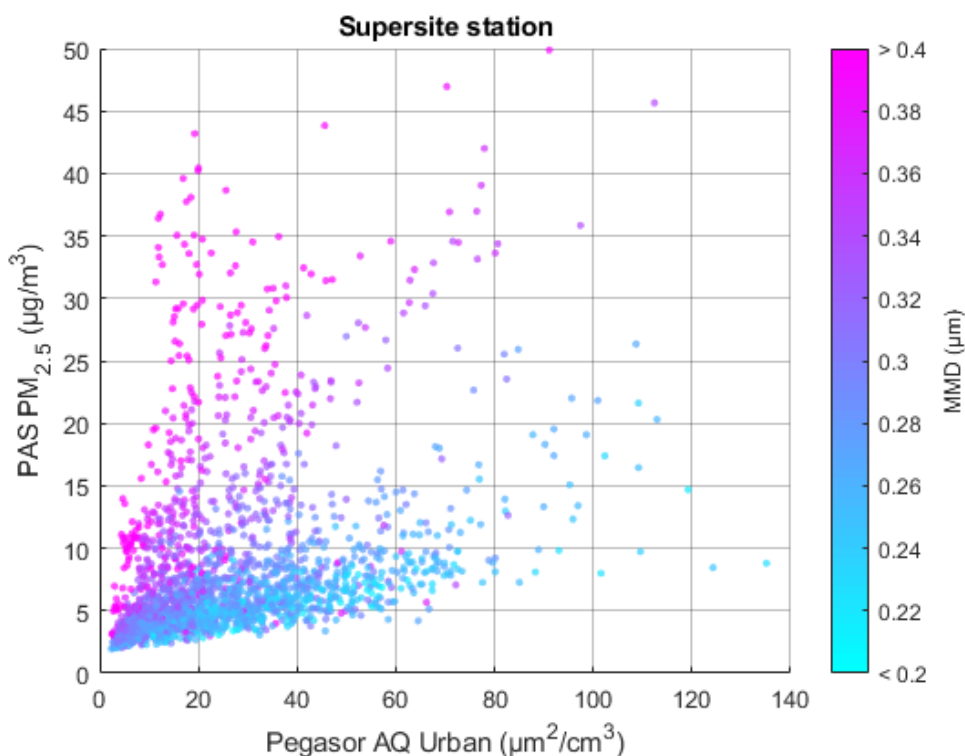

Figure 14. Correlation between the PM2.5 measured with the PAS (Shinyei PPD60PV) and LDSA measured with the Pegasor AQ Urban. The colour indicates the mass median diameter (MMD) calculated from the size distribution data of DMPS.

4.3 Utilization of diffusion charging-based sensors in a monitoring network

The three different urban environments (traffic, urban background, and residential area), in which the measurements of Publication 4 were conducted, showed different types of characteristics regarding measured LDSA concentrations. The year-long mean and standard deviations of LDSA were 22±14, 9.4±6.9, and 12±10 μm² cm^{-3} for the Supersite, UB, and DH1 stations, respectively. Considering the difference between Supersite and UB station, the significance of vicinity traffic is clearly highlighted as these stations were relatively closely located (less than 1 km apart). Globally, the concentrations in Finland

appear to be fairly low. For instance, Ntziachristos et al. (2007) measured concentrations of $53 - 153.4$ µm² cm⁻³ in Los Angeles, Fierz (2011) $11 - 63$ µm² cm⁻³ in Zurich, Buonanno et al. (2012) $69 - 164$ µm² cm⁻³ in Cassino, and Gomes et al. (2012) $35 - 89$ µm² cm⁻³ in Lisbon. The seasonality of the measured LDSA (Figure 15), shows that the concentration levels are highest during summer months (June, July, and August) at all sites in Helsinki Metropolitan area. This is, to some degree, counter-intuitive as traffic rates are typically lower during summer months due to summer holidays, and similarly, residential wood combustion activity is lower due to warmer weather (Helin et al. 2018). It is possible that biogenic precursor gases, which are mainly formed during summer months, enhance secondary organic aerosol (SOA) formation and, being mixed with soot (e.g. BC), increased the size of particles (Zhu et al. 2017). Also, processes governing particle growth can be amplified by stronger radiation and consequent photochemistry (Ma and Birmili 2015; Manninen et al. 2010; Dal Maso et al. 2005). Regardless, this feature of seasonal variation in LDSA concentrations levels hinders slightly the usability of LDSA in local emission source apportionments as it more difficult to judge whether changes in concentrations result from locally emitted direct emission (e.g. residential wood combustion) or from the indirect formation (SOA) of emissions. More research is needed in order to better understand the seasonality of LDSA and its root causes.
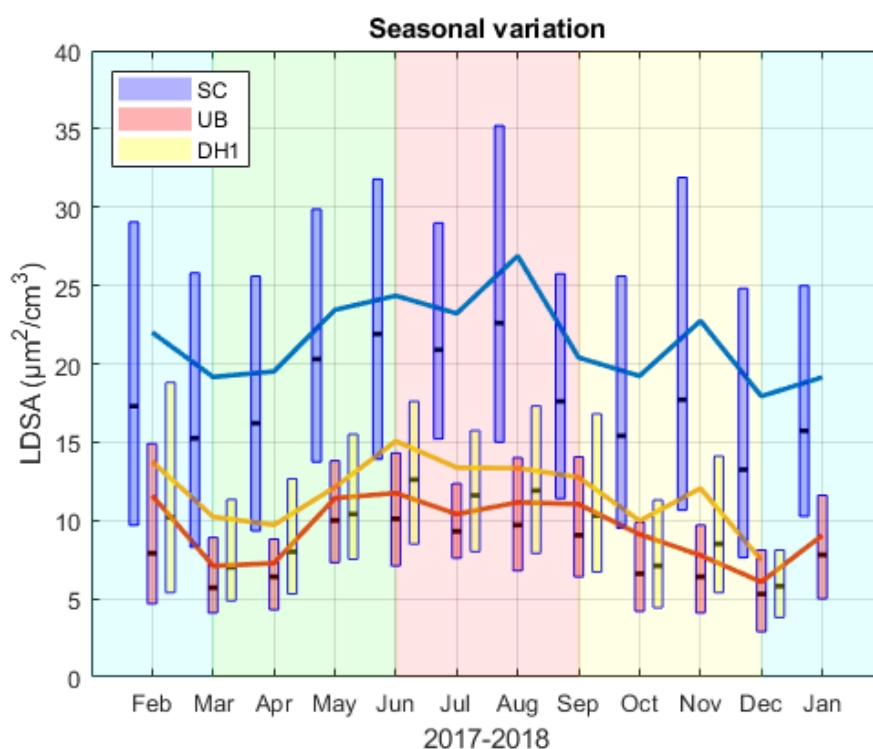
Figure 15. Seasonal variation of LDSA measured at different stations (adapted from Publication 4). Solid continuous line represents monthly mean values, boxes 25th and 75th percentiles and the middle line within the box median values.

Diurnal cycles of LDSA measured at the different stations during winter (December, January, and February) and summer (June, July, and August) are shown in Figure 16. The cycle profiles at Supersite and DH1 stations are similar during both winter and summer seasons; a morning peak is observed at $8 - 9$

am and, for the Supersite station, an afternoon peak at 4 − 5 pm. These are, as stated previously, resulting from the rush hours. For the DH1 station the diurnal cycles have clear seasonal differences. During winter time, the cycle has morning peak at 9 − 10 am and an evening peak at 7 − 9 pm, which are likely associated with morning traffic and evening residential wood combustion. During the summer, however, the cycle profile is bowl-shaped and has no observable day time peaks. This is likely due to the changing boundary layer height which in Finland entails larger day-night variation during summer than during the winter. In summer months, the night time boundary layer height is low which consequently increases the concentrations. During day time, it is the opposite.
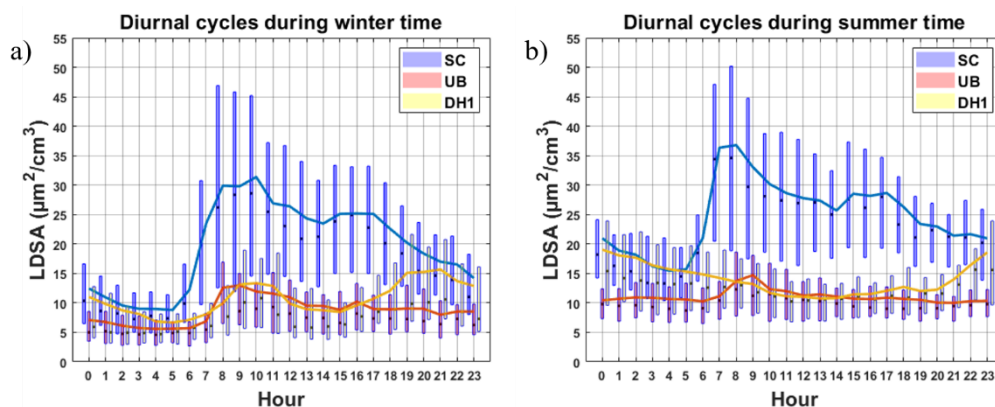
Figure 16. Diurnal cycles at different sites during winter (a) and summer (b) (adapted from Publication 4). Solid continuous lines represent mean values, boxes 25th and 75th percentiles and the line inside the quantile box median values.

Absolute differences between the LDSA measured at DH1 and DH2 stations are shown in Figure 17 where the concentrations are plotted as a function of wind speed (x-axis) and hour of day (colour). The figure shows that significant differences in concentrations are measured despite the proximity of the stations (670 m). The greatest differences are observed in ∼ 225° wind direction and during the evening hours (∼ 3 − 9 pm) which implies that local residential wood burning is the most probable factor causing the differences. A detached housing area forms an environment where multiple sources can be present simultaneously in a relatively small area and thus strong heterogeneity in measured concentrations is possible. This highlights the utility and necessity of a dense monitoring network.

Figure 17. Differences of measured LDSA values at DH1 and DH2 stations as a function of wind direction and hour of day (adapted fr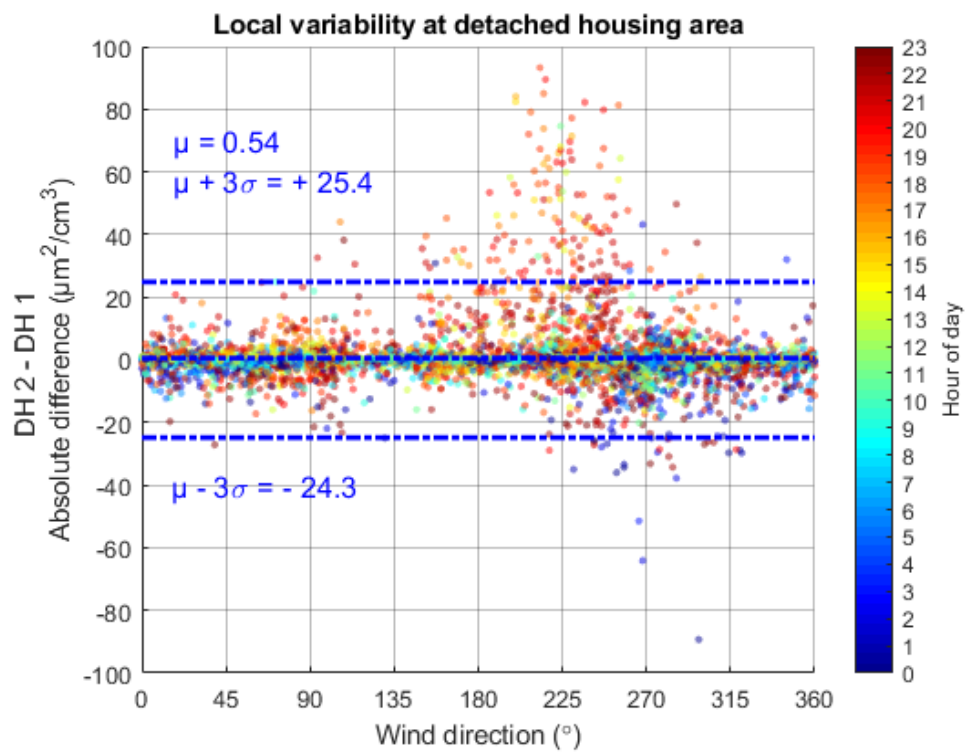om Publication 4). The dashed middle line represents mean and the two dashed-dotted line standard deviations multiplied by 3.

5. Summary and conclusions

Recent emergence and development of aerosol sensors has enabled new possibilities in air quality monitoring. As a result of relatively low unit cost and small size, sensors can be deployed to the field in much higher quantities than conventionally used instruments which have often been perceived as expensive, bulky, and difficult to use. Spatial extension of measurement coverage and the subsequent increase in the understanding of city-scale air quality dynamics is important as particulate matter is one of the leading mortality risk factors in the global burden of disease analysis: several million premature deaths are estimated to occur globally each year due to PM exposure. Distributed sensing could be used to improve, for example, air quality models which, in turn, could be used to developed air quality forecasts and other tools for pollution exposure mitigation. Aerosol sensors could have an essential role in the future when air quality assessments are striving for higher detail and accuracy.

The main parts of this study consisted of a laboratory evaluation of particle size-selectivity of optical aerosol sensors (Publications 1 and 2), field evaluation of the applicability of optical and diffusion charging-based detection techniques to urban air quality monitoring (Publication 3), and of a long-term field measurement of lung deposited surface area in four distinctively different urban environments (Publication 4). Together the publications aimed to provide insight on the accuracy and usability of sensors and demonstrated what kind of benefits sensors could entail when used in a monitoring network application.

The results of Publications 1 and 2 suggest that optical aerosol sensors are unlikely to be fit for long-term regulatory monitoring. The main issues preventing this arise from their improper calibration which poses a significant risk of data misinterpretation; none of the laboratory evaluated sensors measured particle sizes which their technical specifications implied. Although scientific community and experts in the field can measure and quantify the specific response characteristics of a sensor, it is not reasonable to assume that all authorities, who are responsible for the local air quality monitoring, have sufficient facilities and resources available to conduct such experiments. Furthermore, the original idea of a simple and cost-effective complementary monitoring network is easily lost if the used sensors require extensive re-configurations and -calibrations.

With limited operational ranges, optical sensors are suitable to be used in targeted applications (e.g. research-only type) where the characteristics of the sensor response and measured aerosol type are thoroughly known. When considering urban air quality measurements, the sensor accuracy is primarily determined by the valid detection range of the sensor and its comparability to the size distribution of the measured aerosol. As the ambient conditions rarely, if ever, stay stable over longer periods of time, the measurement accuracy will vary accordingly. With known response characteristics, a sensor could be calibrated so that specific aerosol types with known size distribution features are measured accurately (e.g. street dust). However, this would also mean that the proper use of sensor is then limited to the specific conditions only. Related to this, it is worth noting that artificial extension of the sensor operational range using complex statistical models can be unreasonable. Empirical corrections for known artefacts, such as the relative humidity, can be justifiable, however, it is questionable whether data resulting from complex conversion processes (e.g. machine learning) is still a legitimate and independent product of the sensor measurement and not a combination of secondary data and statistical model prediction. Although there is clear evidence showing that machine learning tools can be used to artificially increase the sensor accuracy, problems regarding the metrological characteristics of sensors should still, nevertheless, be solved at the hardware level first.

Despite the discussed issues, optical sensors do entail promising characteristics. Most importantly, the size-selective limitations are not resulting from insurmountable difficulties related to the physics or electronics of optical particle detection (e.g. establishing sufficient signal-to-noise ratio), but rather from their improper configuration. This means that the concept and vision of a sensor driven air quality monitoring network remains valid and achievable. The development of optical sensors should focus on increasing the number size bins, and more importantly, making sure that each size bin is calibrated correctly. Low number of size bins limits the valid operational range and usability of sensors: however, it may be unclear how the amount of advanced measurement features (e.g. multiple size bins) and low unit cost can be reconciled.

The results of Publications 3 and 4 show that the diffusion charging-based detection technique is a reliable and accurate method for the measurement of lung deposited surface area. This observation is in line with the literature, and it indicates that the development and current state of diffusion charger sensors is technologically more mature compared to optical sensors. Lung deposited surface area, as an aerosol metric, is an intermediary parameter which cannot be inferred from either particulate mass or number concentration measurements. However, its usefulness to urban air quality assessments is highlighted by its sensitivity to combustion emitted particles which typically originate from vehicular exhaust emissions and residential wood combustion. As optical methods cannot be used to measure ultrafine particles or LDSA reliably, and particles emitted from combustion sources have a significant contribution to air quality, diffusion charger sensors would be a valuable addition to be included to urban air quality assessments.

The measurements conducted at the three distinctively different urban environments (Publication 4) showed significant differences in site-specific LDSA characteristics despite the relatively close location of stations. For instance, annual concentrations at the Supersite station were more than twice as high as the concentrations at UB station. Likewise, the DH1 and DH2 stations located at the detached housing area showed clearly different diurnal cycles when compared to the diurnal cycles of Supersite and UB stations. High local variability in the measured LDSA concentrations and the distinctive features of different urban environments underlines the need and utility of an air quality monitoring network and higher spatial resolution measurement data.

Future efforts should be focusing on the formation and implementation of uniform classification and testing procedures for aerosol sensors. Currently, the diversity of sensors, their testing methods, and lack of commonly applied metrics and criteria makes the evaluation and inter-comparison of sensors unnecessarily difficult. Formal standardization would give manufacturers, and scientists, a clear goal what to strive for and, consequently, increase the quality of sensors. Improved quality would also encourage air quality authorities to adopt sensors to their monitoring strategies more comprehensively. Besides standardization, another essential factor to consider is the merging of sensor data and air quality models. It is important that the validity and representativeness of the sensors data is pertained so that a model output is not merely a statistical prediction. On the other hand, although the spatial coverage of measurements would ideally be all-encompassing, practical limitations ensure that modelling has an important role in air quality assessments. With proper data fusion, both sensor measurements and models complement each other, and best results are achieved.

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